

**1986 ANNUAL SITE ENVIRONMENTAL REPORT
FOR ARGONNE NATIONAL LABORATORY**

by

N. W. Golchert and T. L. Duffy



ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory (ANL) for 1986 are presented and discussed. To evaluate the effect of Argonne operations on the environment, measurements were made for a variety of radionuclides in air, surface water, ground water, soil, grass, bottom sediment, and milk; of the environmental penetrating radiation dose; and for a variety of chemical constituents in surface water, ground water, and Argonne effluent water. Sample collections and measurements were made on the site, at the site boundary, and off the Argonne site for comparison purposes. The results of the program are interpreted in terms of the sources and origin of the radioactive and chemical substances (natural, fallout, Argonne, and other) and are compared with applicable environmental quality standards. A U. S. Department of Energy (DOE) dose calculation methodology based on recent International Commission on Radiological Protection (ICRP) recommendations is required and used in this report. The radiation dose to off-site population groups is estimated. The average concentrations and total amounts of radioactive and chemical pollutants released by Argonne to the environment were all below appropriate standards.

I. INTRODUCTION

A. General

This report is prepared to provide DOE and the public with information on the levels of radioactive and chemical pollutants in the environment of Argonne and on the amounts, if any, added to the environment as a result of

Argonne operations. The report follows the guidelines given in DOE Draft Order 5484.1A, Chapter III.¹ The Laboratory conducts a continuous monitoring program on and near the Argonne site with the primary purpose of determining the magnitude, origin, and identity of radioactive or chemical substances in the environment. Of special interest is the detection of any such materials released to the environment by Argonne. One important function of the program is to verify the adequacy of Argonne's pollution controls.

Argonne is a DOE energy research and development laboratory with several principal objectives. It conducts a broad program of research in the basic energy and related sciences (physical, chemical, material, nuclear, biomedical, and environmental), and serves as an important engineering center for the study of nuclear and non-nuclear energy sources. Some of the energy-related research projects conducted during 1986 were safety studies for light water and breeder reactors, development of components and materials for fission and fusion reactors, improvements in the utilization of coal for power production (particularly high sulfur coal), electrochemical energy sources including fuel cells and batteries for vehicles and for energy storage, solar energy utilization, evaluation of heat exchangers for the recovery of waste heat from engines, and operation of the National Battery Test Laboratory.

Other areas of research are the use of superconducting magnets for improved nuclear particle accelerators, coal technology, fundamental coal chemistry studies, magnetic confinement fusion, the immobilization of radioactive waste products for safe disposal, medical radioisotope technology, carcinogenesis, and the biological effects of low-level radiation. Environmental research studies include biological activity of energy-related mutagens and carcinogens, chemistry of actinides in natural waters, characterization and monitoring of energy-related pollutants, and the effect of acidic precipitation on vegetation, soil, and surface water quality. A significant portion of these laboratory studies requires the use of radioactive and chemically toxic substances.

The principal nuclear facilities at the Laboratory are a 200 kW light-water cooled and moderated biological research reactor (Janus) fueled with enriched uranium; the Argonne Thermal Source Reactor (ATSR), a 10 kW research reactor fueled with enriched uranium; a superconducting heavy ion linear accelerator (Argonne Tandem Linac Accelerating System, ATLAS); a 22 MeV pulsed electron linac; a 60-inch cyclotron; several other charged particle accelerators (principally of the Van de Graaff and Dynamitron type); a large fast neutron source (Intense Pulsed Neutron Source, IPNS) in which high energy protons strike a heavy metal target to produce the neutrons; cobalt-60 irradiation sources; chemical and metallurgical plutonium laboratories; and hot cells and laboratories designed for work with multicurie quantities of the actinide elements and with irradiated reactor fuel elements.

Two experiments, initiated in 1984, were continued in these facilities in 1986 and have some potential environmental impact: 1) proof-of-breeding in light-water reactors, which involves the dissolution and analysis of irradiated thorium and uranium-233 dioxide fuel elements and 2) recovery of tritium from reactor irradiated ceramic lithium compounds. Two major facilities, a 12.5 GeV proton accelerator (Zero Gradient Synchrotron, ZGS) and a 5 MW heavy water enriched uranium reactor (CP-5) were shut down several years ago. The CP-5 facility is awaiting decontamination and decommissioning (D&D), while the D&D work has been completed at the ZGS.

The DOE New Brunswick Laboratory, a safeguards plutonium and uranium measurements and analytical chemistry laboratory, is located on the ANL site. The principal non-nuclear activities at Argonne that may produce a measurable impact on the environment are a coal-fired boiler (No. 5), the closed-loop heat exchanger studies for waste heat recovery, and the Fossil Energy Users Laboratory (FEUL). The boiler is designed to burn high sulfur (3.5%) Illinois coal to produce export steam for Laboratory use. It is equipped with a slaked lime spray scrubber and bag collector to reduce sulfur dioxide and particulate emissions. The heat recovery studies use moderately large quantities of toxic or flammable organic compounds such as toluene, Freon, biphenyl oxides, methyl pyridine, and trifluoroethanol. The

major potential environmental impact of these materials would be accidental release due to equipment malfunction.

B. Description of Site

Argonne National Laboratory (Illinois site) occupies the central 688 hectares (1,700 acres) of a 1514-hectare (3,740-acre) tract in DuPage County, 43 km (27 mi) southwest of downtown Chicago, and 39 km (24 mi) due west of Lake Michigan. It lies in the Des Plaines River Valley, south of Interstate Highway 55 and west of Illinois Highway 83. Figures 1 and 2 are maps of the site and of the surrounding area. The 826-hectare (2,040-acre) area surrounding the site (Waterfall Glen Forest Preserve) was formerly Argonne property, but was deeded to the DuPage County Forest Preserve District in 1973 for use as a public recreational area, nature preserve, and demonstration forest.

The terrain is gently rolling, partially-wooded, former prairie and farmland. The grounds contain a number of small ponds and streams, the principal one being Sawmill Creek, which runs through the site in a southerly direction and enters the Des Plaines River about 2.1 km (1.3 mi) southeast of the center of the site. The land is drained primarily by Sawmill Creek, although the extreme southern portion drains directly into the Des Plaines River, which flows along the southern boundary of the Forest Preserve. This river flows southwest until it joins the Kankakee River about 48 km (30 mi) southwest of the Laboratory to form the Illinois River.

The largest topographical feature is the Des Plaines River channel, about 1.6 km (1 mi) wide. This channel contains the River, the Chicago Sanitary and Ship Canal, and the Illinois and Michigan Canal. Their presence extends the uninhabited area about 1.6 km (1 mi) south of the site. The elevation of the channel surface is 180 m (590 ft) above sea level. Bluffs, which comprise the south border of the site, rise from the channel at varying slope angles of 15° to 60°, reaching an average elevation of 200 m (650 ft) above sea level at the top. The land then slopes gradually upward reaching the average site elevation of 220 m (725 ft) above sea

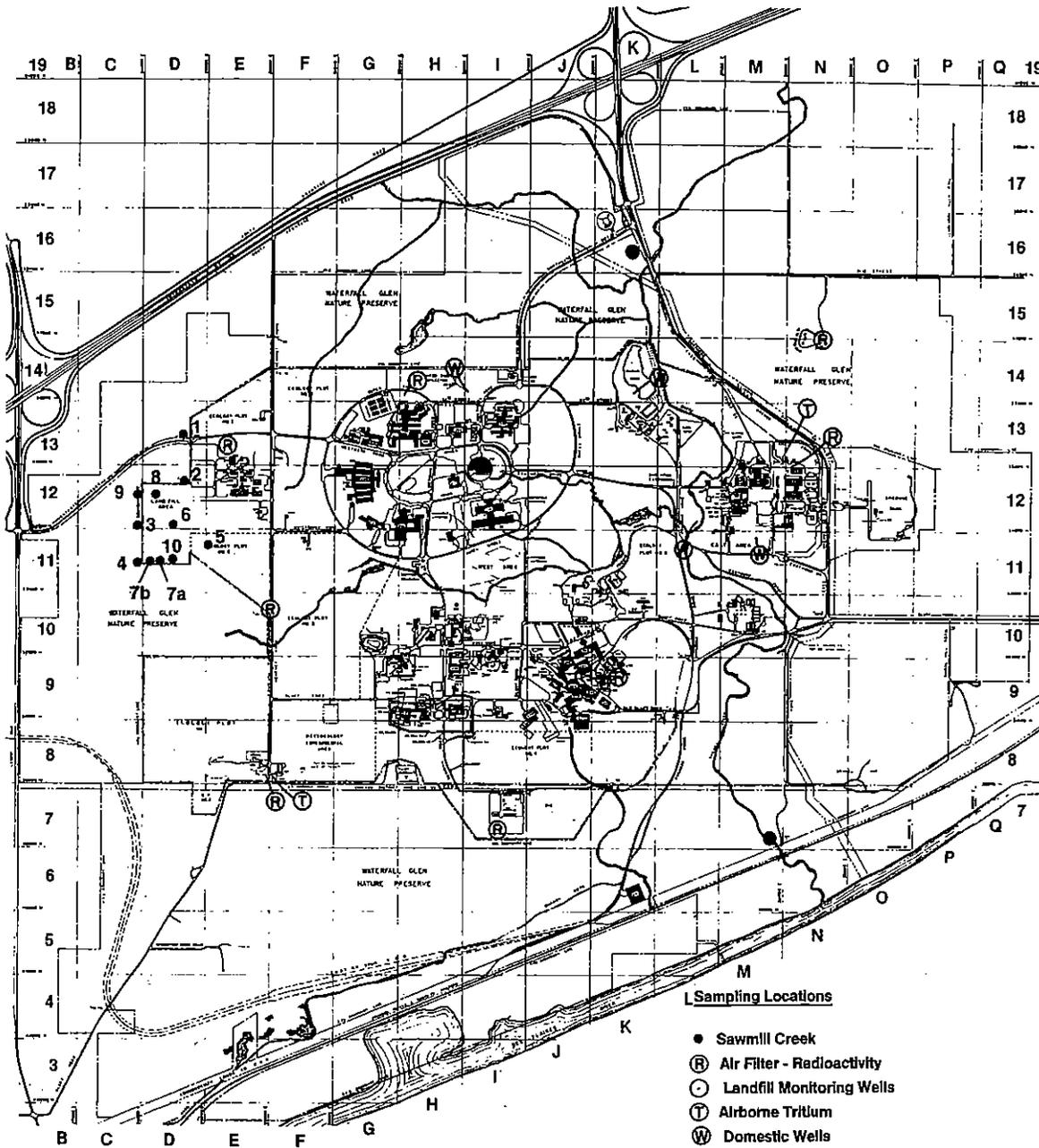


Fig. 1. Sampling Locations at Argonne National Laboratory

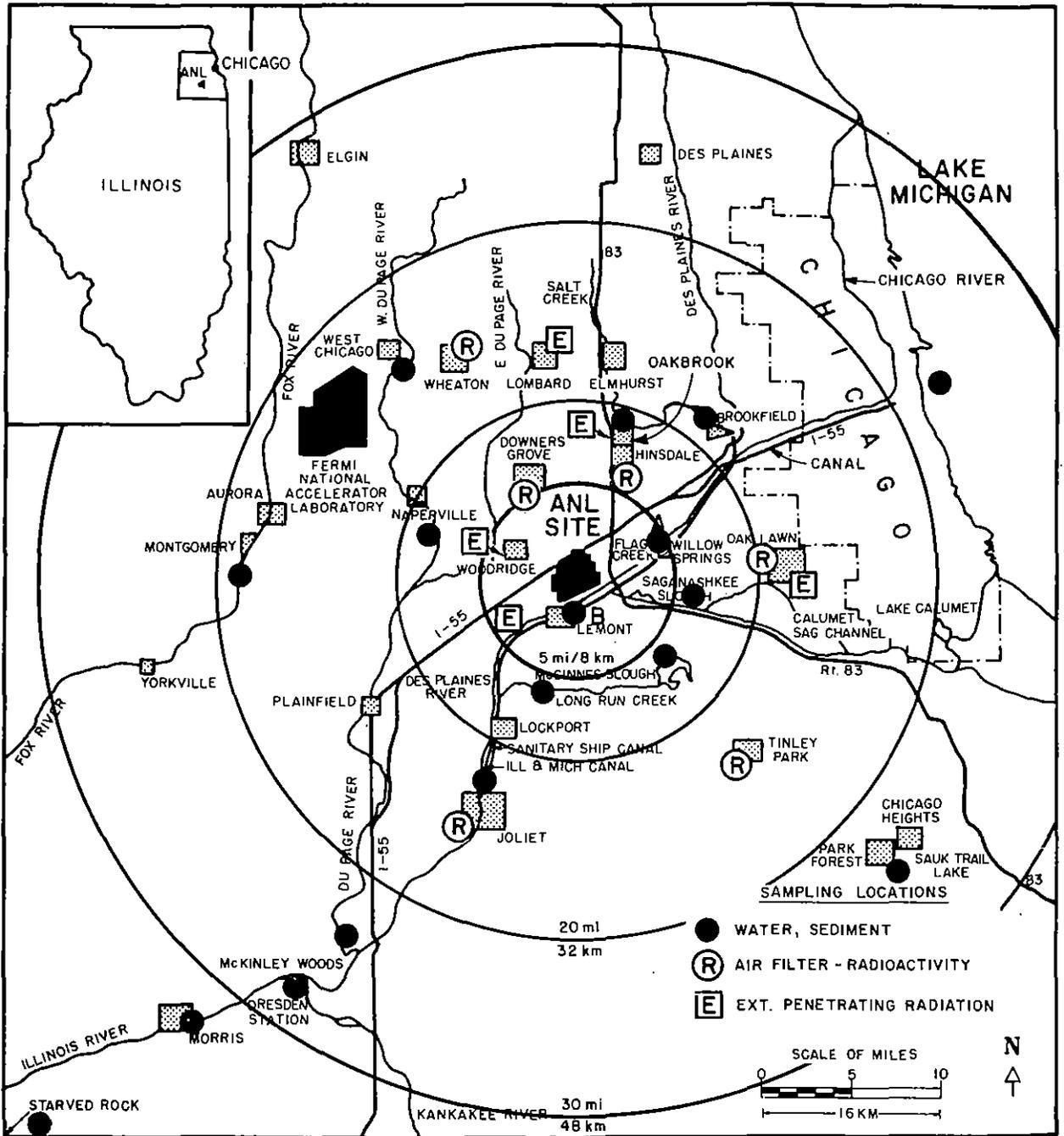


Fig. 2. Sampling Locations Near Argonne National Laboratory

level at 940 m (3,000 ft) from the bluffs. Several large ravines oriented in a north-south direction are located in the southern portion of the site. The bluffs and ravines generally are forested with mature deciduous trees. The remaining portion of the site changes in elevation by no more than 7.6 m (25 ft) in a distance of 150 horizontal m (500 ft). In the southern portion of the Forest Preserve, the Chicago District Pipe Line Co. and the Atchison, Topeka, and Santa Fe Railroad have rights-of-way. Additional information about the site is given in the Argonne Environmental Assessment.²

C. Population

The area around Argonne has exhibited a large population growth in the past 30 years. Large areas of farmland have been converted into housing. A directional and annular 80-km (50-mi) population distribution for the area, which is used for the population dose calculations later in this report, is shown in Table 1. The population distribution was obtained by modifying a distribution for 1981 prepared by Urban Decision Systems, Inc. and which was based on the 1980 census. The values for distances within 8 km (5 mi) of the site were modified by using quarter-section population data supplied by the Northeastern Illinois Planning Commission, as adjusted on the basis of local observations.

D. Climatology

The climate of the area is that of the upper Mississippi Valley, as moderated by Lake Michigan. A summary of the meteorological data collected on the site from 1950 to 1964 is available³ and provides a historical sample of the climate.

The most important meteorological parameters for the purposes of this report are wind direction, wind speed, temperature, and precipitation. The wind data are used to calculate radiation doses from air emissions and in selecting air sampling locations and distances. Temperature and precipitation data are useful in interpreting some of the environmental results. Because on-site meteorological data are no longer available in suitable form,

TABLE 1

Incremental Population Data in the Vicinity of ANL, 1981

Distance, miles Distance, km	In Thousands									
	0 - 1 0-1.6	1 - 2 1.6-3.2	2 - 3 3.2-4.8	3 - 4 4.8-6.4	4 - 5 6.4-8.0	5-10 8-16	10-20 16-32	20-30 32-48	30-40 48-64	40-50 64-80
<u>Direction</u>										
N	0	344	1504	863	4115	37.2	179.2	312.1	133.3	202.1
NNE	0	188	2086	14685	5882	38.8	290.7	493.4	95.9	0
NE	0	528	6544	1450	1219	44.0	710.1	940.7	0	0
ENE	0	2630	3640	1854	985	35.6	630.5	240.8	0	0
E	0	14	212	20	15	34.4	514.9	249.4	10.7	25.2
ESE	0	0	85	275	120	11.3	206.2	291.9	271.0	69.0
SE	0	5	155	225	68	29.0	69.5	119.2	24.4	13.3
SSE	0	44	2299	1422	120	1.9	21.7	9.3	9.2	20.0
S	0	100	574	2114	725	5.5	18.5	1.8	33.0	39.5
SSW	0	60	4407	1928	705	19.1	100.9	9.4	17.7	7.5
SW	0	620	1304	50	915	13.1	31.5	6.5	15.0	7.8
WSW	0	492	50	409	12261	3.3	7.1	2.1	6.3	9.4
W	0	2853	905	14000	16464	4.1	58.7	19.6	15.0	6.6
WNW	0	1007	140	5100	5960	39.8	85.5	8.7	7.7	50.3
NW	0	215	2032	3367	7741	28.5	65.2	87.2	10.5	16.6
NNW	0	323	987	2156	7710	41.1	151.2	167.1	107.7	79.5
Total	0	9423	26924	49918	65005	386.7	3141.4	2959.2	757.4	546.8
Cumulative Total	0	9423	36347	86265	151270	538.0	3679.4	6638.6	7396.0	7942.8

measurements taken in 1986 at the nearest U. S. Weather Bureau station, O'Hare Airport, 31 km (18 mi) north of Argonne, were used. The 1986 average monthly and annual wind roses are shown in Figure 3. The wind roses are polar coordinate plots in which the lengths of the radii represent the percentage frequency of wind speeds in classes of 2.01-6 m/s (4.5-13.4 mph), 6.01-10 m/s (13.4-22.4 mph), and greater than 10.01 m/s (22.4 mph). The number in the center represents the percent of observations of wind speed less than 2 m/s (4.5 mph) in all directions. The direction of the radii from the center represents the direction from which the wind blows. Sixteen radii are shown on each plot at 22.5° intervals; each radius represents the average wind speed for the direction covering 11.25° on either side of the radius.

A comparison of the monthly wind roses indicates that the winds are sufficiently variable that monitoring for airborne releases must be carried out in all directions from the site. For example, the dominant wind direction in March is south-southwest while in June it is northeast. The annual average wind rose for 1986 is consistent with the long term average wind direction, which usually varies from the west to south, but with a significant northeast component. Precipitation and temperature data for 1986 are shown in Table 2. The monthly precipitation data for 1986 was marked by large differences from the average. For example, February, September, and October were significantly above the average, while January, March, April, August, and December were below normal. However, the annual total was only about 5% lower than the average. The temperatures were about average throughout the year.

E. Geohydrology

The geology of the Laboratory area consists of about 30 m (100 ft) of glacial till overlying dolomite bedrock. The bedrock is the Niagaran and Alexandrian dolomite of Silurian age. These formations are underlain by Maquoketa shale of Ordovician age, and older dolomites and sandstones of Ordovician and Cambrian age. The beds are nearly horizontal.

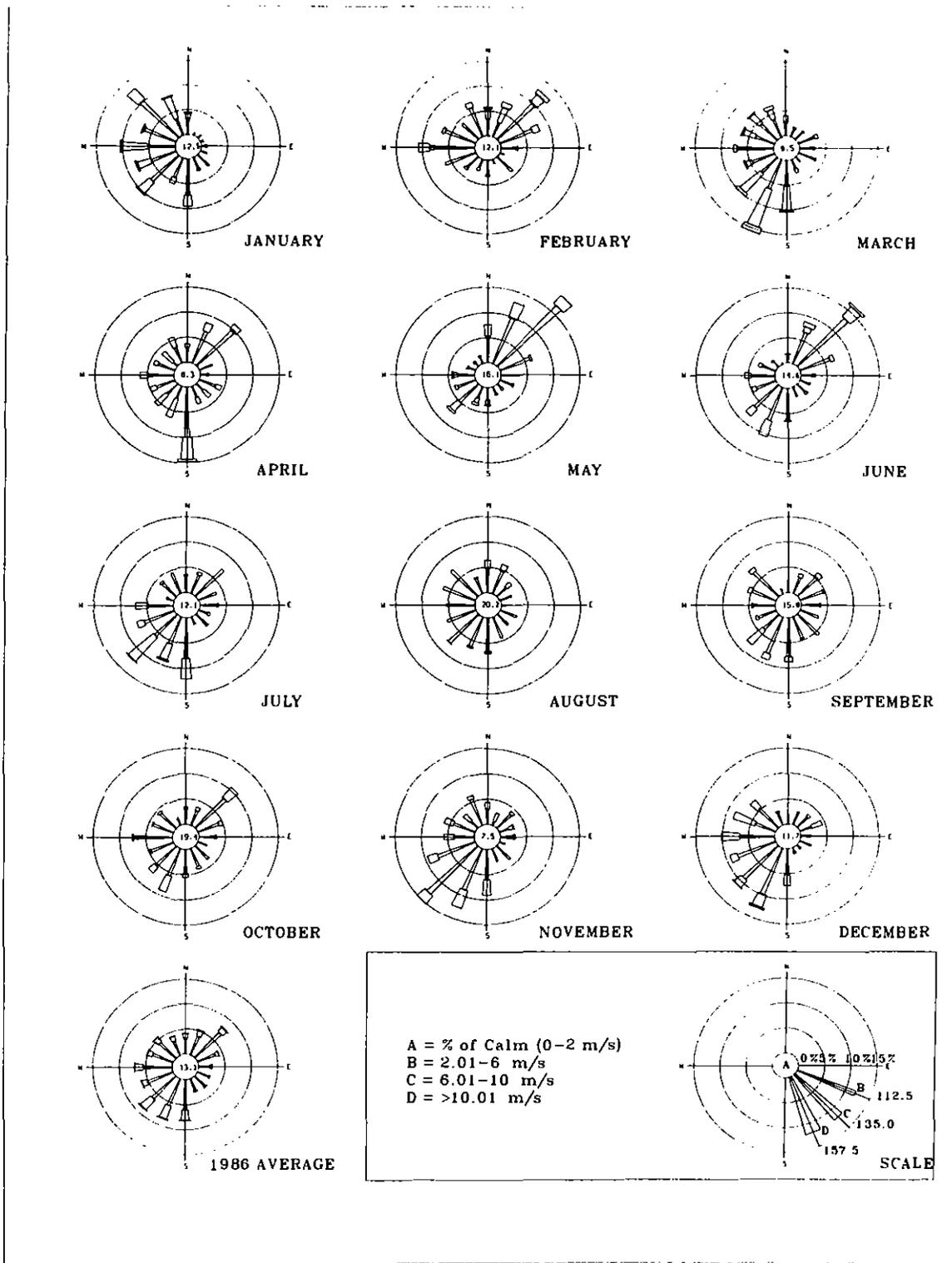


Fig. 3. Monthly and Annual Wind Roses Near Agonne National Laboratory, 1986

TABLE 2

ANL Weather Summary, 1986*

Month	Precipitation (cm)		Temperature (°C)	
	Amount	Average	Monthly Average	Average
January	0.99	4.06	-5.1	-5.9
February	6.55	3.33	-4.4	-3.3
March	3.78	6.58	4.7	2.2
April	4.70	9.30	10.8	9.3
May	7.90	8.00	15.3	15.1
June	8.86	10.36	19.1	20.3
July	10.92	9.22	23.8	22.8
August	2.92	8.97	20.3	22.2
September	18.08	8.51	19.3	18.2
October	9.53	5.79	12.1	11.9
November	3.58	5.23	2.2	4.3
December	2.77	5.33	-0.8	-2.4
Total	80.58	84.68		

* Data obtained from the National Oceanic and Atmospheric Administration (NOAA) for the weather station at O'Hare International Airport. The average is for the years 1951-1980.

Two principal aquifers are used as water supplies in the vicinity of the Laboratory. The upper aquifer is the Niagaran-Alexandrian dolomite, which is about 60 m (200 ft) thick in the Laboratory area, and has a piezometric surface between 15 and 30 m (50 and 100 ft) below the ground surface over much of the site. The lower aquifer is the Galesville sandstone, which lies between 150 and 450 m (490 and 1,500 ft) below the surface. The Maquoketa shale separates the upper dolomite aquifer from the underlying sandstone aquifer. This shale retards hydraulic connection between the upper and lower aquifers.

The four domestic water wells now in use are about 90 m (300 ft) deep in the Niagaran dolomite. One well, in the Galesville sandstone 490 m (1,600 ft) deep, is not used because the water table has dropped below the pumping level. The water level in the Niagaran dolomite has remained reasonably stable under Laboratory pumping, dropping about 3.7 m (12 ft) between 1960 and 1980. The aquifer appears to be adequate for future Laboratory use, however, this ground water source is also used throughout the area. There are several monitoring wells and small capacity water wells used for laboratory experiments, fire protection, and sanitary facilities.

F. Water and Land Use

The principal stream that drains the site is Sawmill Creek. It carried effluent water continuously from a sewage treatment plant (Marion Brook Treatment Plant) located a few kilometers north of the site until October 27, 1986, when the plant was closed. The plant had an operating capacity of about 11.4 megaliters (3 million gallons) per day but frequently operated significantly above its capacity. In addition, the residential development in the area has resulted in the collection and channeling of additional runoff water into the Creek. Treated sanitary and laboratory waste water from Argonne are combined and discharged into Sawmill Creek at location 7M in Figure 1. This effluent averaged 2.8 megaliters (0.74 million gallons) per day. The combined Argonne effluent consisted of 52% laboratory waste water and 48% sanitary waste water. The water flow in the Creek upstream of the waste-water outfall averaged about 31 megaliters

(8.2 million gallons) per day during 1986. Before the Marion Brook Treatment Plant closed on October 27, 1986, the Creek flow averaged 34 megaliters (9.1 million gallons) per day while after closure, the flow average was 17 megaliters (4.5 million gallons) per day.

Sawmill Creek and the Des Plaines River above Joliet, about 21 km (13 mi) southwest of Argonne, receive very little recreational or industrial use. A few people fish in these waters downstream from Argonne and some duck hunting takes place on the Des Plaines River. Water from the Chicago Sanitary and Ship Canal is used by Argonne for cooling towers and by others for industrial purposes, such as hydroelectric generators and condensers, and for irrigation at the State prison near Joliet. The Argonne usage is about 0.4 megaliter (100,000 gallons) per day. The Canal, which receives Chicago Metropolitan Sanitary District effluent water, is used for industrial transportation and some recreational boating. Near Joliet, the River and Canal are combined into one waterway, which continues until it joins the Kankakee River to form the Illinois River about 48 km (30 mi) southwest of Argonne. The Dresden Nuclear Power Station complex is located at the confluence of the Kankakee, Des Plaines, and Illinois Rivers. This Station uses water from the Kankakee River for cooling, and discharges the water into the Illinois River. The first use of water for drinking is an indirect one at Alton, on the Mississippi River about 710 km (370 mi) downstream from Argonne, where water is used to replenish ground water supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from both shallow and deep aquifers) and Lake Michigan water are used for drinking purposes.

The principal recreational area near Argonne is Waterfall Glen Forest Preserve, which surrounds the site as described in Section I.B. and is shown in Figure 1. The area is available for hiking, skiing, and equestrian sports. Sawmill Creek flows through the south portion of the Preserve on its way to the Des Plaines River. Several large forest preserves of the Cook County Forest Preserve District are located east and southeast of Argonne and the Des Plaines River. The preserves include the two sloughs shown in Figure 2, McGinnis and Saganashkee, as well as other smaller lakes. These areas are used for picnicking, boating, fishing, and

hiking. A small park located in the eastern portion of the Argonne site (Location 12-0 in Figure 1) is for the use of Argonne and Department of Energy employees only.

II. SUMMARY

This is one in a series of annual reports prepared to provide DOE, environmental agencies, and the public with information on the level of radioactive and chemical pollutants in the environment and on the amounts of such substances, if any, added to the environment as a result of Argonne operations. Included in this report are the results of measurements obtained in 1986 for a number of radionuclides in air, surface water, ground water, soil, grass, bottom sediment, and milk; for the external penetrating radiation dose; and for a variety of chemical constituents in surface and subsurface water. The previous report in this series is ANL-86-13.⁴

The major airborne radionuclides released from the Laboratory were hydrogen-3, carbon-11, argon-41, krypton-85, and radon-220, all as gases. The maximum dose from these nuclides at the site boundary was 0.44 mrem/y in the north direction, as calculated from an atmospheric dispersion model. The calculated dose to the closest full-time resident, who is located about 0.5 km (0.3 mi) north of the site boundary, was 0.045 mrem/y, which is 0.045% of the DOE 100 mrem/y limit for prolonged exposures. The hazard due to a given concentration of radionuclide or quantity of external radiation is assessed in this report by calculating the corresponding effective dose equivalent and comparing it to the DOE recommended dose limits discussed in Section III.A. and III.A.7., and described in References 5 and 6. These releases constitute an insignificant addition to the dose received from the natural background radiation, which is about 78 mrem/y. The total 80-km population dose from these radionuclides was less than 2.9 man-rem for 1986, compared to approximately 6.2×10^5 man-rem from the natural background radiation.

Radioactivity on airborne particulates was measured in air-filter samples collected continuously at the site perimeter and off the site. The filters were analyzed for total alpha, total beta, fission and activation products, thorium, uranium, and plutonium. No activity attributable to Argonne operations could be detected. These samples contained only radionuclides from natural sources and nuclear test detonations. Increased

concentrations of several volatile fission products were observed in May, June, and July as a result of fallout from the Chernobyl accident.

Argonne waste water is discharged into Sawmill Creek, and this stream was sampled above and below the site to evaluate the effect of Argonne operations on its radioactive content. The nuclides (for which analyses were made) added to the Creek in the waste water, and the ingestion dose from their net average Creek concentrations, if the Creek water had been used as a potable water supply, were hydrogen-3, 0.116 mrem/y; strontium-90, 0.328 mrem/y; neptunium-237, 0.0003 mrem/y; plutonium-239,240, 0.0006 mrem/y; and americium-241, 0.018 mrem/y. The concentrations and corresponding doses are all very low compared to the 100 mrem/y dose limit.

Sawmill Creek flows into the Des Plaines River, which in turn flows into the Illinois River. The radioactivity levels in the Rivers were similar to those in other streams in the area, and the activity added to the Creek by Argonne waste water had no measurable effect on the radioactive content of either the Des Plaines or Illinois Rivers.

Plutonium concentrations in soil showed the same general range and average at the site perimeter and off the site as in the past years. The average plutonium-239,240 content of the top 5 cm (2 in) of soil was $1.2 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ at the site perimeter and $0.8 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ off the site. The corresponding plutonium-238 averages were $0.03 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ at both locations. The plutonium content of grass was similar to that found in previous years and was about a factor of 10^4 less than soil from the same locations. The results were within the range reported by other laboratories for fallout from test detonations, and the plutonium found in soil and grass is attributed to this source. The plutonium content of samples from beds of off-site streams and ponds ranged from $1 \times 10^{-9} \mu\text{Ci}/\text{g}$ to $20 \times 10^{-9} \mu\text{Ci}/\text{g}$ of plutonium-239,240, a range found in previous years to be normal for fallout plutonium in such materials. However, concentrations twice normal were found in the sediment just below the Laboratory waste-water outfall as a result of their presence in Argonne waste water.

Milk from a dairy farm near the Laboratory was analyzed for hydrogen-3 and strontium-90. Hydrogen-3 concentrations averaged $< 100 \times 10^{-9}$ $\mu\text{Ci/mL}$. The strontium-90 concentration of 2.8×10^{-9} $\mu\text{Ci/mL}$ was similar to the 1985 result. These radionuclides resulted from nuclear test detonations, and are not related to Argonne operations.

Measurements of external penetrating radiation were made at several locations at the site boundary and off the site. The off-site results averaged 78 ± 5 mrem/y, which is in the normal range for the area. At two site boundary locations, above-normal readings were recorded that were attributable to Argonne operations. At the south fence (grid 7I in Figure 1), the dose rate averaged about 366 mrem/y above normal as a result of radiation from an on-site temporary storage facility for radioactive waste. About 300 m (0.2 mi) south of the fence, the measured dose rate decreased to 78 ± 9 mrem/y, which is within the normal range. Along the north side of the site, the dose at the fence at location 14I was 24 mrem/y above normal due to radiation from cobalt-60 sources in Building 202. Since all of these locations are unoccupied, there are no individuals receiving these measured doses. The calculated outdoor dose rate from these sources to the residents closest to the south boundary, about 1.6 km (1 mi) from the fence line, was about 0.08 mrem/y, which is 0.08% of the DOE dose limit. Similarly, the dose rate to the residents closest to the north boundary, about 0.75 km (0.5 mi) from the fence, was about 0.11 mrem/y, which is 0.11% of the dose limit.

Concentrations of chemical constituents and other water quality parameters were measured in Argonne waste and effluent water and in Sawmill Creek. The results were compared to the standards adopted by the State of Illinois as well as National Pollutant Discharge Elimination System (NPDES) permit limits.

All the results obtained for the ANL waste water were below the State standards except for mercury, which exceeded the State standard in two samples. The average mercury concentration was 29% of the State effluent standard. Efforts are continuing to lower these concentrations by additional treatment of batches of waste water that contain elevated levels of mercury.

The results for the NPDES permit locations were generally in compliance with the exception of the pH at Location 009 (Fig. 6). This remained above the limit through November, due to the basicity of water from the water softening process, but it no longer flows since a new softening process has been instituted and water now discharges at Location 001.

The average values in Sawmill Creek for dissolved oxygen and most chemical constituents were within the State of Illinois Water Quality Standards. The average levels of copper and iron were 85% and 70% of the State standards, and individual values exceeded the standards 23% and 15% of the time, respectively. Copper and iron concentrations increased in the stream as a result of turbidity due to rainfall runoff. Elevated levels of these elements in the Creek are believed to be of natural origin. Mercury concentrations exceeded the State standard on one occasion, and the average concentration was 19% of the standard. The concentration of mercury in the Des Plaines River was not affected by the amounts in Argonne effluent water.

Additional monitoring wells were constructed at the sanitary landfill to expand and improve the coverage. Samples from these monitoring wells were examined for selected inorganic and organic constituents. Levels of manganese were elevated in two wells, the pH was elevated in a third well, and arsenic was elevated in another. Similar results were obtained in the past, and there is no indication of migration between wells since the relative concentrations have not changed with time. Samples were examined for 13 organic compounds and none were detected.

The average concentrations and total amounts of radioactive and chemical pollutants released by Argonne to the environment did not constitute a health hazard. Any individual discharges from the Laboratory that exceeded acceptable standards were temporary, and when they did occur, investigations were undertaken to identify the source and reduce their discharge.

III. MONITORING RESULTS

A. Radiological

The radioactivity of the environment was determined by measuring the concentrations of radioactive nuclides in naturally-occurring materials and by measuring the external radiation dose. Sample collections and measurements were made at the site perimeter and off the site principally for comparison purposes. Some on-site results are also reported when they are useful in interpreting perimeter and off-site results. Since radioactivity is usually transported by air and water, the sample collection program has concentrated on these media. In addition, soil, plants, foodstuffs, and materials from the beds of lakes and streams were also collected and analyzed. The program followed the guidance provided in the DOE Environmental⁷ and Effluent⁸ Surveillance Guides. About 1600 samples were collected and approximately 4000 analyses were performed.

The results of radioactivity measurements are expressed in terms of microcuries per milliliter ($\mu\text{Ci/mL}$) for water, air, and milk, and microcuries per gram ($\mu\text{Ci/g}$) and/or microcuries per square meter ($\mu\text{Ci/m}^2$) for soil, bottom sediment, and vegetation. When a nuclide was not detected, the result is given as less than (<) the minimum amount detectable (detection limit) by the analytical method used. Averages, including individual results that were less than the detection limit, were calculated by one of the following two methods. If a large fraction (usually 50% or more) of the individual results was less than the detection limit, the average was calculated with the assumption that such results were equal to the detection limit, and the resulting average value is expressed as less than (<) the computed average. If only a small fraction of the individual results was less than the detection limit, the average was calculated with the assumption that such results were actually one-half of the detection limit, and the average is given as a definite value. The former technique probably overestimates the average concentration in those samples below the detection limit and gives an upper limit for the average of all the samples in the group, since it is unlikely that all concentrations not detectable are at the detection limit. The latter method is based on the assumption

that the values below the detection limit are distributed between zero and the detection limit with a frequency such that the average value is one-half of the detection limit. The averages that are obtained by using these two methods under the conditions indicated are believed to give an adequate picture of the average concentration at locations where the concentrations not only varied greatly, but were at times not detectable. Penetrating radiation measurements are reported in units of millirem (mrem) per year and population dose in man-rem.

Average values are usually accompanied by a plus-or-minus (+) limit value. Unless otherwise stated, this value is the standard error at the 95% confidence level calculated from the standard deviation of the average, and is a measure of the range in the concentrations encountered at that location. It does not represent the conventional uncertainty in the average of repeated measurements on the same or identical samples. Since many of the variations observed in environmental radioactivity are not random but occur for specific reasons (e.g., seasonal variations), samples collected from the same location at different times are not replicates. The more random the variation in activity at a particular location, the closer the confidence limits will represent the actual distribution of values at that location. The averages and confidence limits should be interpreted with this in mind. When a plus-or-minus figure accompanies an individual result in this report, it represents the statistical counting error at the 95% confidence level.

DOE has provided interim guidance⁶ for dose equivalent calculations for members of the public based on ICRP-26 and ICRP-30.⁵ These procedures have been used in this report and it is expected that this approach will replace the Concentration Guides (CGs) in DOE Order 5480.1A, Chapter XI, that have been used in the past to compare environmental radionuclide concentrations with DOE standards.⁹ The new methodology requires three components to be calculated: 1) the committed dose equivalent from all sources of ingestion, 2) the committed dose equivalent from inhalation, and 3) direct dose equivalent from external radiation. These three components are summed for comparison with the new DOE dose equivalent limits for environmental exposure. The interim guidance requires that sufficient data be

available on exposure to radionuclides and sources to assure that at least 90% of the total committed effective dose equivalent is accounted for. The primary radiation dose limit for members of the public is 500 mrem/y for occasional annual exposures and 100 mrem/y for prolonged exposures (greater than five years). The effective dose equivalents for members of the public from all routine DOE operations, natural background and medical exposures excluded, shall not exceed these values. Routine DOE operations are normally planned operations, which exclude actual or potential accidental or unplanned releases.

The measured or calculated environmental radionuclide concentrations or radiation dose is converted to a 50-year committed effective dose equivalent with the use of the Effective Dose Equivalent Factors (EFF.D.E.) and compared to the annual dose limits for uncontrolled areas. The EFF.D.E.s and annual dose limits are both given in the interim guidance.⁶ The numerical values of the EFF.D.E.s used in this report are given in Section III.A.7. Although the EFF.D.E.s apply only to dose rates from concentrations above natural levels, the calculated dose is sometimes given in this report for activities that are primarily of natural origin for comparison purposes. Such values are enclosed in parentheses to indicate this. Occasionally other standards are used and their source is identified in the text.

1. Air

The radioactive content of particulate matter was determined by collecting and analyzing air-filter samples. The sampling locations are shown in Figures 1 and 2. Separate particulate collections were made for specific radiochemical analyses. Alpha, beta, and gamma measurements were made on samples collected continuously on laminated glass fiber filters which were changed weekly at eight locations at the Argonne site perimeter and at five locations off the site. The site perimeter samplers are placed at the nearest location to the site boundary fence that provides electrical power and shelter. Comparison between perimeter and off-site concentrations is necessary to evaluate and establish the Laboratory contribution. If only off-site radioactivity were reported, their normality or origin could not

be evaluated. Higher activities at the site perimeter may indicate radioactivity released by Argonne if the differences are greater than the error in sampling and measurement. Such results require investigation to determine the cause of the difference. The relative error is between 5 and 20% for most results, but approaches 100% at the detection limit.

The total alpha and beta activities in the individual weekly samples are summarized in Table 3. These measurements were made in low-background gas-flow proportional counters, and the counting efficiencies used to convert counting rates to disintegration rates were those measured for radon decay products on filter paper. The average concentrations of gamma-ray emitters, as determined by gamma-ray spectrometry performed on composite weekly samples are given in Table 4. The gamma-ray detector is a shielded germanium diode calibrated for each gamma-ray emitting nuclide measured.

The alpha activity, principally due to naturally occurring nuclides, averaged the same as the past several years and was in its normal range. Excluding May and June, the perimeter beta activity averaged 2.7×10^{-14} $\mu\text{Ci/mL}$, which is the same as the average value for the past four years. The increased beta activities in May and June resulted from the Chernobyl accident and are discussed below. The gamma-ray emitters listed in Table 4 are those that have been present in the air for the past few years and are of natural origin. The beryllium-7 exhibits a spring increase in concentration, indicating its stratospheric origin. The lead-210 in air is due to the radioactive decay of gaseous radon-222.

An accident at a U.S.S.R. nuclear power facility, Chernobyl, occurred on Saturday, April 26, 1986. Airborne radioactivity from this accident was identified in a number of European countries. In anticipation of the eventual spread of the radioactivity to this country, daily air-filter samples were collected and measured by gamma-ray spectrometry. Fallout from the accident was first identified in a sample collected on the weekend of May 9 to May 12, 1986. The highest concentrations were found in the air sample of May 14, 1986, and a second smaller peak was observed on May 22, 1986.

TABLE 3
 TOTAL ALPHA AND BETA ACTIVITIES IN AIR-FILTER SAMPLES, 1986*
 (CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	NO. OF SAMPLES	ALPHA ACTIVITY			BETA ACTIVITY		
			AV.	MIN.	MAX.	AV.	MIN.	MAX.
JANUARY	PERIMETER	28	2.2	1.0	7.9	28	19	38
	OFF-SITE	15	1.9	0.4	3.2	27	16	42
FEBRUARY	PERIMETER	31	2.0	0.7	2.9	29	19	40
	OFF-SITE	17	1.9	1.1	3.1	27	18	38
MARCH	PERIMETER	30	1.8	0.6	3.8	22	15	35
	OFF-SITE	18	1.8	0.8	2.6	18	12	26
APRIL	PERIMETER	39	2.4	1.3	4.3	23	12	35
	OFF-SITE	24	2.2	0.9	3.9	19	8	32
MAY	PERIMETER	32	1.7	0.6	2.8	146	15	349
	OFF-SITE	19	1.5	0.3	2.9	137	18	299
JUNE	PERIMETER	32	2.2	1.1	4.0	67	19	185
	OFF-SITE	17	1.9	0.8	3.7	61	6	180
JULY	PERIMETER	36	2.3	0.1	7.9	25	14	49
	OFF-SITE	25	2.4	0.4	9.1	23	9	46
AUGUST	PERIMETER	27	2.8	1.4	7.0	30	20	41
	OFF-SITE	18	1.8	0.6	2.9	25	12	41
SEPTEMBER	PERIMETER	31	2.0	0.8	7.2	26	7	41
	OFF-SITE	17	1.4	0.2	3.0	24	5	50
OCTOBER	PERIMETER	37	1.5	0.1	3.8	24	8	49
	OFF-SITE	18	1.4	0.6	3.6	22	8	59
NOVEMBER	PERIMETER	26	2.2	1.4	4.0	30	21	40
	OFF-SITE	14	1.9	0.9	4.6	27	12	43
DECEMBER	PERIMETER	25	1.9	1.1	3.2	29	20	41
	OFF-SITE	19	2.1	0.6	4.4	28	10	60
ANNUAL SUMMARY	PERIMETER	374	2.1 ± 0.1	0.1	7.9	40 ± 4	7	349
	OFF-SITE	221	1.8 ± 0.1	0.2	9.1	37 ± 4	5	299

* THESE RESULTS WERE OBTAINED BY MEASURING THE SAMPLES FOUR DAYS AFTER THEY WERE COLLECTED TO AVOID COUNTING THE NATURAL ACTIVITY DUE TO SHORT-LIVED RADON AND THORON DECAY PRODUCTS. THIS ACTIVITY IS NORMALLY PRESENT IN THE AIR AND DISAPPEARS WITHIN FOUR DAYS BY RADIO-ACTIVE DECAY.

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NOTE: (1E-15 = 10⁻¹⁵)

TABLE 4

GAMMA RAY ACTIVITY IN AIR-FILTER SAMPLES, 1986
(CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	BE7	PB210
JANUARY	PERIMETER	77	36
	OFF-SITE	83	39
FEBRUARY	PERIMETER	63	39
	OFF-SITE	71	48
MARCH	PERIMETER	92	23
	OFF-SITE	90	25
APRIL	PERIMETER	128	20
	OFF-SITE	117	24
MAY	PERIMETER	96	19
	OFF-SITE	100	23
JUNE	PERIMETER	139	22
	OFF-SITE	104	22
JULY	PERIMETER	127	26
	OFF-SITE	127	36
AUGUST	PERIMETER	131	32
	OFF-SITE	134	35
SEPTEMBER	PERIMETER	91	31
	OFF-SITE	82	31
OCTOBER	PERIMETER	70	28
	OFF-SITE	89	36
NOVEMBER	PERIMETER	68	36
	OFF-SITE	61	31
DECEMBER	PERIMETER	63	39
	OFF-SITE	51	41
ANNUAL SUMMARY	PERIMETER	95 ± 17	29 ± 4
	OFF-SITE	93 ± 15	33 ± 5
DOSE(REM)	PERIMETER	(0.00022)	(0.032)
	OFF-SITE	(0.00021)	(0.036)

The concentrations then decreased until August, after which no fission products from the accident could be detected. The results of the daily samples are collected in Table 5.

The radionuclides in Table 5 are characterized as being volatile or having volatile precursors, with iodine-131 as the dominant species. Parallel sample collections with paper and charcoal cartridges indicated that essentially all of the iodine was associated with particulate matter. In addition to the daily samples, the normal weekly composited air-filter papers were gamma-ray counted and the larger volume allowed the identification of ruthenium-106-rhodium-106 and cesium-136 in the air. The composite samples were used to measure the gamma-ray emitters identified in Table 5 until near the end of July when all concentrations were less than the detection limit of 1×10^{-16} $\mu\text{Ci/mL}$.

The inhalation dose, using the methodology in Section III.7.b, was calculated for the gamma-ray emitting nuclides listed in Table 5. A summation of the daily doses calculated by this method would give 0.007 mrem. This is very small compared to the dose received from natural sources, approximately 4.3 mrem over the same time period, and implies that the impact from the releases of this accident should have no measurable impact on the population in this area.

Samples for radiochemical analyses were collected at perimeter locations 12N and 7I (Figure 1) and off the site in Downers Grove (Figure 2). Collections were made on polystyrene filters. The total air volume filtered for the monthly samples was about 25,000 m^3 . Samples were ignited at 600°C to remove organic matter and prepared for analysis by vigorous treatment with hot hydrochloric, hydrofluoric, and nitric acids.

Plutonium and thorium were separated on an anion exchange column and the uranium was extracted from the column effluent. Following the extraction, the aqueous phase was analyzed for radiostrontium by a standard radiochemical procedure. The separated plutonium, thorium, and uranium fractions were electrodeposited and measured by alpha spectrometry. The chemical recoveries were monitored by adding known amounts of plutonium-242,

TABLE 5

Major Gamma-Ray Emitting Radionuclides in Air from the Chernobyl Accident
(Concentrations in 10^{-15} $\mu\text{Ci/mL}$)

Collection Time On	Collection Time Off	Ruthenium-103	Iodine-131	Cesium-134	Cesium-137	Barium-140- Lanthanum-140	Inhalation* Dose During Sampling Period (mrem)
(0824)5/9	(0823)5/12	10 \pm 2	314 \pm 24	7 \pm 1	20 \pm 2	< 1	0.00025
(0826)5/12	(0815)5/13	72 \pm 6	1809 \pm 80	70 \pm 6	141 \pm 9	< 1	0.0015
(0818)5/13	(0820)5/14	123 \pm 7	2154 \pm 86	76 \pm 6	169 \pm 10	28 \pm 7	0.0018
(0824)5/14	(0810)5/15	98 \pm 7	865 \pm 60	37 \pm 5	91 \pm 8	18 \pm 7	0.00076
(0812)5/15	(0812)5/16	37 \pm 5	302 \pm 41	16 \pm 3	34 \pm 5	6 \pm 4	0.00027
(0815)5/16	(0818)5/19	23 \pm 2	193 \pm 15	9 \pm 1	23 \pm 2	< 1	0.00017
(0821)5/19	(0817)5/20	20 \pm 3	362 \pm 37	11 \pm 3	31 \pm 9	< 1	0.00030
(0819)5/20	(0817)5/21	95 \pm 6	848 \pm 54	40 \pm 4	79 \pm 7	< 1	0.00075
(0820)5/21	(0810)5/22	291 \pm 10	1258 \pm 68	54 \pm 5	127 \pm 9	14 \pm 5	0.0011
(0813)5/22	(0814)5/23	70 \pm 5	289 \pm 34	10 \pm 3	48 \pm 6	< 1	0.00027
(0816)5/23	(0831)5/27	28 \pm 1	14 \pm 1	7 \pm 1	12 \pm 1	< 1	0.00030
(0834)5/27	(0818)5/28	13 \pm 3	6 \pm 3	3 \pm 2	3 \pm 2	< 1	0.000013
(0821)5/28	(0852)5/29	8 \pm 3	5 \pm 3	6 \pm 3	4 \pm 3	< 1	0.000015

*See Section III.7.b.

thorium-229, and uranium-232 tracers prior to ignition. Since alpha spectrometry cannot distinguish between plutonium-239 and plutonium-240, it should be understood that when plutonium-239 is mentioned in this report, the alpha activity due to the plutonium-240 isotope is also included. The results are given in Table 6.

Measurable strontium-89 was detected in the May and June samples and is attributed to fallout from the Chernobyl accident. The May and June strontium-90 results were also slightly elevated but the average concentration is similar to that of 1985. The average plutonium-239 concentration is slightly less than last year and there is no evidence of any plutonium-239 due to the Chernobyl accident. Figure 4 shows the monthly plutonium-239 off-site air concentrations for the past 14 years. The arrows indicate the approximate dates of atmospheric nuclear tests. An examination of Figure 4 indicates that plutonium in air exhibits significant increases in concentration in the spring following an atmospheric nuclear test. The magnitude of the increase is related to the size of the test.

The thorium and uranium concentrations are in the same range found during the past several years and are considered to be of natural origin. The amounts of thorium and uranium in a sample were proportional to the mass of inorganic material collected on the filter paper. The bulk of these elements in the air was due to resuspension of soil. In contrast, the amount of plutonium in the air samples contributed by resuspended soil ranged from about 5% to 76% and averaged 25% of the total plutonium in the samples. This assumes that the resuspended soil has the same plutonium concentration as the first centimeter on the ground. The remainder of the plutonium-239 is due to world-wide fallout.

The major airborne effluents released during 1986 are listed in Table 7 by location. The increase from Building 200, as compared to 1985, is due to work on the "proof-of-breeding" program and the hydrogen-3 from Building 212 is from the tritium recovery studies. In addition to the nuclides listed in Table 7, several other fission products were also released in millicurie or smaller amounts. The quantities listed in Table 7 were measured by in-line stack monitors in the exhaust systems of the buildings.

TABLE 6
STRONTIUM, THORIUM, URANIUM AND PLUTONIUM CONCENTRATIONS IN AIR-FILTER SAMPLES, 1986
(CONCENTRATIONS IN $1\text{E}-18$ MICROCURIES/HL)

MONTH	LOCATION ¹	2							2		3
		SR-89	SR-90	TH-228	TH-230	TH-232	U-234	U-235	U-238	PU-239	
JANUARY	7I	< 100	27 ± 22	7 ± 2	11 ± 1	5 ± 1	8 ± 1	0.8 ± 0.6	8 ± 1	1.0 ± 0.3	
	12N	< 100	< 10	18 ± 9	25 ± 5	15 ± 4	39 ± 2	1.8 ± 0.7	38 ± 2	1.2 ± 0.3	
	OFF-SITE	< 100	< 10	7 ± 2	12 ± 3	3 ± 2	22 ± 3	< 0.3	21 ± 3	0.8 ± 0.6	
FEBRUARY	7I	< 100	13 ± 5	2 ± 1	6 ± 1	3 ± 1	10 ± 1	< 0.3	11 ± 1	0.7 ± 0.3	
	12N	< 100	< 10	9 ± 1	15 ± 1	8 ± 1	18 ± 1	< 0.3	19 ± 1	0.6 ± 0.2	
	OFF-SITE	< 100	< 10	3 ± 1	5 ± 1	2 ± 1	7 ± 1	< 0.3	8 ± 1	0.4 ± 0.2	
MARCH	7I	< 100	< 10	5 ± 1	7 ± 1	4 ± 1	8 ± 1	< 0.3	7 ± 1	0.6 ± 0.3	
	12N	< 100	< 10	14 ± 1	22 ± 1	14 ± 1	21 ± 2	< 0.3	19 ± 2	0.6 ± 0.2	
	OFF-SITE	< 100	12 ± 8	3 ± 2	6 ± 1	3 ± 1	9 ± 1	< 0.3	6 ± 1	0.7 ± 0.3	
APRIL	7I	< 100	19 ± 24	12 ± 1	33 ± 2	8 ± 1	16 ± 2	< 0.3	14 ± 1	0.4 ± 0.3	
	12N	< 100	< 10	11 ± 1	22 ± 1	12 ± 1	20 ± 2	< 0.3	23 ± 2	0.6 ± 0.3	
	OFF-SITE	< 100	14 ± 3	5 ± 2	7 ± 1	4 ± 1	9 ± 1	< 0.3	8 ± 1	0.4 ± 0.2	
MAY	7I	3500 ± 2400	56 ± 40	6 ± 3	9 ± 1	6 ± 1	11 ± 1	< 0.3	12 ± 1	0.7 ± 0.3	
	12N	2600 ± 460	13 ± 7	6 ± 3	11 ± 1	6 ± 1	15 ± 1	< 0.3	13 ± 1	0.7 ± 0.3	
	OFF-SITE	2400 ± 780	13 ± 13	4 ± 4	5 ± 1	4 ± 1	11 ± 1	< 0.3	10 ± 1	0.4 ± 0.2	
JUNE	7I	810 ± 270	52 ± 40	12 ± 2	16 ± 1	5 ± 1	9 ± 1	< 0.3	8 ± 1	0.7 ± 0.3	
	12N	940 ± 190	14 ± 5	11 ± 1	19 ± 1	12 ± 1	15 ± 1	< 0.3	14 ± 1	0.8 ± 0.3	
	OFF-SITE	240 ± 90	39 ± 13	7 ± 2	10 ± 1	3 ± 1	8 ± 1	< 0.3	5 ± 1	0.7 ± 0.3	
JULY	7I	< 100	< 10	8 ± 2	9 ± 1	5 ± 1	5 ± 1	< 0.3	6 ± 1	1.1 ± 0.3	
	12N	< 100	< 10	20 ± 7	44 ± 5	19 ± 2	12 ± 1	0.4 ± 0.3	12 ± 1	1.6 ± 0.7	
	OFF-SITE	< 100	< 10	7 ± 3	12 ± 2	3 ± 1	5 ± 1	< 0.3	5 ± 1	0.5 ± 0.2	
AUGUST	7I	< 100	< 10	27 ± 4	71 ± 4	18 ± 2	5 ± 1	< 0.3	6 ± 1	3.0 ± 0.8	
	12N	< 100	< 10	5 ± 4	12 ± 2	5 ± 1	10 ± 2	0.7 ± 0.4	11 ± 2	1.9 ± 0.6	
	OFF-SITE	< 100	< 10	3 ± 2	8 ± 2	3 ± 1	11 ± 1	0.5 ± 0.3	10 ± 1	0.7 ± 0.3	
SEPTEMBER	7I	< 100	15 ± 4	17 ± 3	24 ± 2	15 ± 2	9 ± 1	< 0.3	6 ± 1	0.6 ± 0.3	
	12N	< 100	22 ± 6	7 ± 3	14 ± 2	6 ± 1	12 ± 2	< 0.3	11 ± 1	0.5 ± 0.3	
	OFF-SITE	< 100	23 ± 3	2 ± 2	13 ± 1	1 ± 1	6 ± 1	< 0.3	4 ± 1	0.5 ± 0.3	
OCTOBER	7I	< 100	< 10	7 ± 3	9 ± 3	5 ± 2	8 ± 1	< 0.3	6 ± 1	0.4 ± 0.3	
	12N	< 100	< 10	4 ± 2	7 ± 1	4 ± 1	10 ± 1	0.4 ± 0.3	10 ± 1	0.6 ± 0.4	
	OFF-SITE	< 100	< 10	4 ± 1	5 ± 1	1 ± 1	4 ± 1	< 0.3	4 ± 1	0.4 ± 0.3	
NOVEMBER	7I	< 100	< 10	3 ± 2	12 ± 3	7 ± 2	16 ± 2	< 0.3	8 ± 2	0.6 ± 0.3	
	12N	< 100	< 10	20 ± 3	38 ± 3	19 ± 1	21 ± 3	< 0.3	18 ± 3	0.9 ± 0.4	
	OFF-SITE	< 100	27 ± 31	1 ± 2	5 ± 1	2 ± 1	6 ± 1	< 0.3	4 ± 1	0.3 ± 0.3	
DECEMBER	7I	< 100	15 ± 7	5 ± 1	8 ± 2	2 ± 1	9 ± 1	< 0.3	9 ± 1	0.5 ± 0.3	
	12N	< 100	11 ± 12	9 ± 1	15 ± 1	5 ± 1	16 ± 2	< 0.3	15 ± 1	0.3 ± 0.3	
	OFF-SITE	< 100	16 ± 15	3 ± 1	4 ± 1	2 ± 1	7 ± 1	< 0.3	6 ± 1	0.4 ± 0.3	
ANNUAL	7I	< 440	20 ± 10	9 ± 4	18 ± 11	7 ± 3	9 ± 2	< 0.3	8 ± 2	0.9 ± 0.4	
	12N	< 380	< 10	11 ± 3	20 ± 7	10 ± 3	17 ± 5	< 0.5	17 ± 5	0.9 ± 0.3	
SUMMARY	OFF-SITE	< 300	10 ± 10	4 ± 1	8 ± 2	3 ± 1	9 ± 3	< 0.3	7 ± 3	0.5 ± 0.1	
DOSE (MREH) (X 1E-5)	7I	< 13.7	22	(0.24)	(0.92)	(0.006)	(.10)	(< 0.003)	(0.09)	0.024	
	12N	< 11.8	< 11	(0.29)	(1.05)	(0.009)	(.19)	(< 0.005)	(0.17)	0.024	
	OFF-SITE	< 9.33	11	(0.11)	(0.40)	(0.002)	(.09)	(< 0.003)	(0.08)	0.014	

¹ PERIMETER LOCATIONS ARE GIVEN IN TERMS OF THE GRID COORDINATES IN FIGURE 1.

² THE CONCENTRATIONS IN UNITS OF MICROGRAMS/CUBIC METER CAN BE OBTAINED BY MULTIPLYING THE VALUE IN MICROCURIES/HL BY 2.96×10^{12} FOR URANIUM-238 AND BY 9×10^{12} FOR THORIUM-232. THE MASS OF THE OTHER THORIUM ISOTOPES IN COMPARISON TO THORIUM-232 AND THE OTHER URANIUM ISOTOPES IN COMPARISON TO URANIUM-238 IS NEGLIGIBLE.

³ PLUTONIUM-240 IS INCLUDED (SEE TEXT).

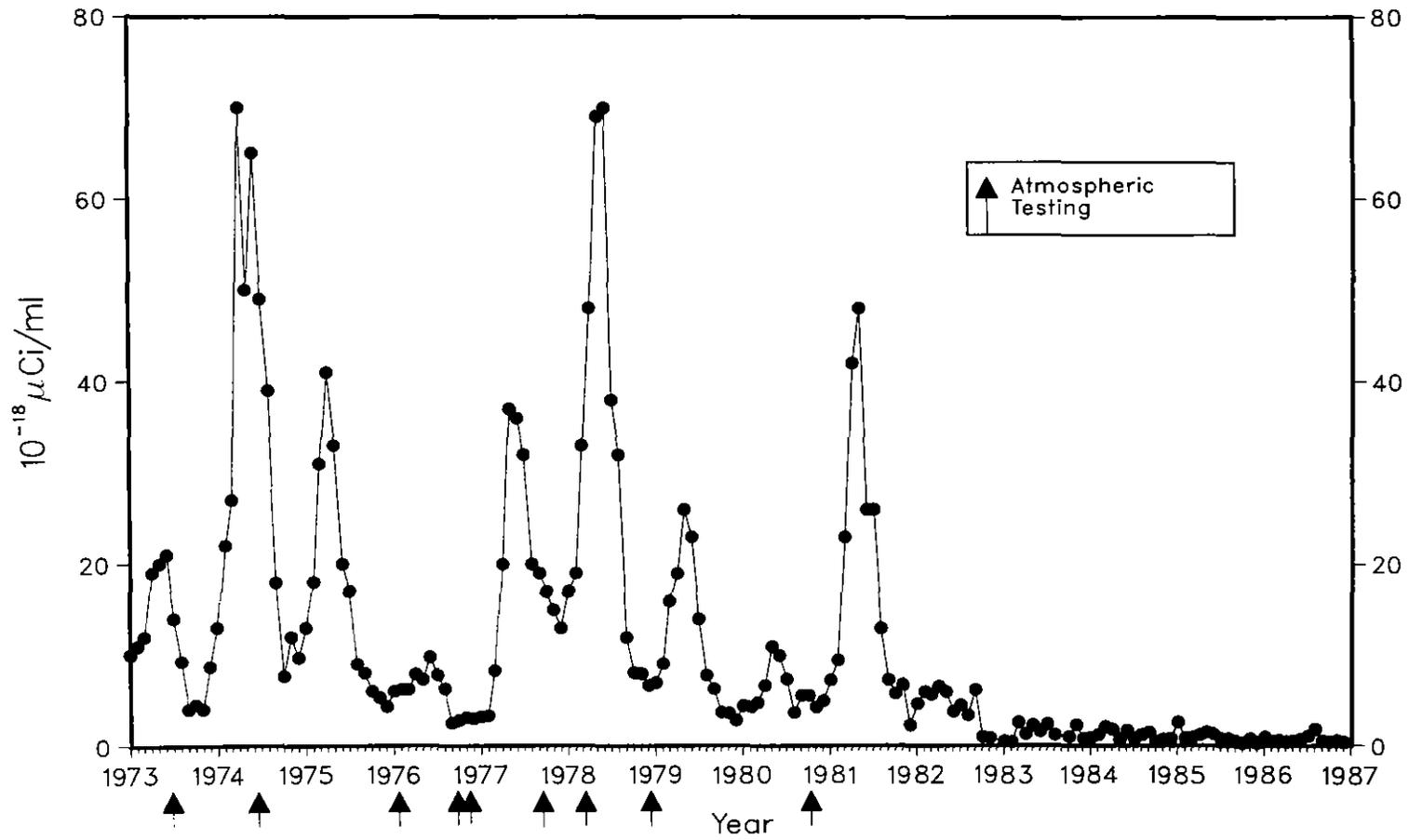


Fig. 4. Plutonium-239,240 Air Concentrations, 1973-1986

TABLE 7

Summary of Airborne Radioactive Emissions, 1986

Building	Nuclide	Half-Life	Amount Released (Curies/year)
200	Hydrogen-3	12.3 y	0.9
	Radon-220	56 s	6982
202 (Janus)	Argon-41	1.8 h	1.5
212	Hydrogen-3	12.3 y	10.3
	Krypton-85	10.7 y	1.7
331 (CP-5)	Hydrogen-3	12.3 y	40
375 (IPNS)	Carbon-11	20 m	90

An air sampling program for determining tritium in air was carried out because the Laboratory is conducting a program that could result in the release of tritiated water vapor into the air. Samples were collected at perimeter locations, 8F (at the southwest corner of the site) and at 12N (on the east perimeter of the site), and off the site in Woodridge, Illinois. The tritiated water vapor was collected by adsorption on silica gel and was measured by counting the desorbed water in a liquid scintillation spectrometer. The results are given in Table 8. Based on the data in Table 7, the principal sources of the tritiated water vapor should be from Building 212, Location 12I and Building 330, Location 9H (CP-5). Because the winds are usually from the west to south, the tritium concentrations should be higher at equal distances east and north of the release points. However, the concentrations at 8F were almost twice as high as at 12N, because this location is closer to the principal source (CP-5). At all sampling locations the doses were very low.

2. Surface Water

Total (nonvolatile) alpha and beta activities were determined by counting the residue remaining after evaporation of the water, and applying

TABLE 8

Tritiated Water Vapor in Air, 1986
(Concentrations in 10^{-12} $\mu\text{Ci/mL}$)

Month	Location*	No. of Samples	Avg.	Min.	Max.
January	8F	8	0.20	< 0.1	0.45
	12N	8	0.64	0.42	0.91
February	8F	8	1.99	0.35	4.96
	12N	8	0.67	0.16	2.57
March	8F	9	0.44	< 0.1	1.05
	12N	9	0.58	< 0.1	2.41
April	8F	9	1.26	< 0.1	3.92
	12N	9	0.41	< 0.1	1.28
	Off-Site	2	0.21	0.11	0.31
May	8F	9	1.99	0.14	5.05
	12N	9	0.37	< 0.1	0.87
	Off-Site	2	0.30	0.29	0.31
June	8F	8	2.40	0.24	5.41
	12N	8	1.17	0.15	2.45
	Off-Site	2	1.04	0.56	1.51
July	8F	9	3.70	0.89	9.05
	12N	9	2.04	0.20	5.84
	Off-Site	2	0.50	0.24	0.75
August	8F	9	2.24	0.59	5.70
	12N	9	1.34	0.19	3.11
	Off-Site	1	-	-	1.53
September	8F	8	0.41	0.14	1.05
	12N	8	0.72	0.20	1.55
	Off-Site	1	-	-	0.60
October	8F	9	1.74	0.13	5.97
	12N	9	0.73	0.25	1.57
	Off-Site	1	-	-	0.26
November	8F	9	0.23	< 0.1	1.00
	12N	9	0.29	< 0.1	0.62
	Off-Site	2	0.12	< 0.1	0.19
December	8F	6	0.44	< 0.1	1.81
	12N	6	1.03	< 0.1	3.53
	Off-Site	2	0.34	0.32	0.37
Annual Summary	8F	101	1.42	< 0.1	9.05
	12N	101	0.83	< 0.1	5.84
	Off-Site	15	0.49	< 0.1	1.53
Dose (mrem)	8F	-	0.00075	< 0.00005	0.0048
	12N	-	0.00044	< 0.00005	0.0031
	Off-Site	-	0.00026	< 0.00005	0.00081

* Locations are given in terms of the grid coordinates in Figure 1.

counting efficiency corrections determined for uranium-233 (for alpha activity) and thallium-204 (for beta activity) to obtain disintegration rates. Hydrogen-3 was determined on a separate sample, and this activity does not appear in the total nonvolatile beta activity. Uranium was determined using a laser fluorometer, and the results calculated in terms of activity with the assumption that the isotopic composition was that of natural uranium. Analyses for other radionuclides were performed by specific radiochemical separations followed by appropriate counting. One liter aliquots were used for all analyses except hydrogen-3 and the transuranium nuclides. Hydrogen-3 analyses were performed by liquid scintillation counting of 10 mL in a gel system. Analyses for transuranium nuclides were performed on 10-liter samples by chemical separation methods followed by alpha spectrometry.^{10,11} Plutonium-236 was used to determine the yields of plutonium and neptunium, which were separated together. A group separation of a fraction containing the transplutonium elements was monitored for recovery with americium-243 tracer.

Argonne waste water is discharged into Sawmill Creek, a small stream that runs through the Laboratory grounds, drains surface water from much of the site, and flows into the Des Plaines River about 500 m (0.3 mi) downstream from the waste-water outfall. Sawmill Creek was sampled upstream from the Argonne site and downstream from the waste-water outfall to determine if radioactivity was added to the stream by Argonne waste water or surface drainage. The sampling locations are shown in Figure 1. Below the waste-water outfall, daily samples were collected by a continuous sampler, which operated about 62% of the year. Construction activities in the Creek caused most of the 38% downtime. When the continuous sampling device was not functioning, a grab sample was collected each working day. Equal portions of the daily samples collected each week were combined and analyzed to obtain an average weekly concentration. Above the site, samples were collected once a month and were analyzed for the same radionuclides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 9. Comparison of the results, and 95% confidence levels of the

TABLE 9
RADIOMUCLIDES IN SAWHILL CREEK WATER, 1986

TYPE OF ACTIVITY	LOCATION *	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/HL)			AVG.	DOSE (HREM)	
			AVG.	MIN.	MAX.		MIN.	MAX.
ALPHA (NONVOLATILE)	16K	12	2.2 ± 0.1	1.4	3.9	-	-	-
	7H	248	1.7 ± 0.1	1.0	2.9	-	-	-
BETA (NONVOLATILE)	16K	12	20 ± 1	7	35	-	-	-
	7H	248	32 ± 16	9	345	-	-	-
HYDROGEN-3	16K	12	< 125	< 100	277	< 0.0057	< 0.005	0.0128
	7H	248	2650 ± 3985	< 100	103100	0.1219	< 0.005	4.7426
STRONTIUM-90	16K	12	< 0.28	< 0.25	0.46	< 0.027	< 0.02	0.04
	7H	248	3.74 ± 3.52	< 0.25	78.3	0.355	< 0.02	7.43
URANIUM (NATURAL)	16K	12	1.0 ± 0.1	0.5	1.8	(0.18)	(0.079)	(0.30)
	7H	248	0.9 ± 0.1	0.4	2.1	(0.16)	(0.067)	(0.35)
NEPTUNIUM-237	16K	12	< 0.001	< 0.001	0.002	< 0.0003	< 0.00028	0.00055
	7H	248	0.0020 ± 0.0007	< 0.001	0.014	0.0006	< 0.00028	0.0041
PLUTONIUM-238	16K	12	-	-	< 0.001	-	-	< 0.00028
	7H	248	< 0.0012	< 0.001	0.004	0.00032	< 0.00028	0.00099
PLUTONIUM-239	16K	12	-	-	< 0.001	-	-	< 0.00031
	7H	248	0.0028 ± 0.0009	< 0.001	0.0176	0.0009	< 0.00031	0.0055
AMERICIUM-241	16K	12	-	-	< 0.001	-	-	< 0.0016
	7H	248	0.0122 ± 0.0051	< 0.001	0.096	0.0195	< 0.0016	0.154
CURIUM-242 AND/OR CALIFORNIUM-252	16K	12	-	-	< 0.001	-	-	< 0.0003
	7H	248	< 0.0012	< 0.001	0.0027	< 0.0004	< 0.0003	0.0010
CURIUM-244 AND/OR CALIFORNIUM-249	16K	12	-	-	< 0.001	-	-	< 0.0017
	7H	248	< 0.0020	< 0.001	0.0111	< 0.0033	< 0.0017	0.0186

* LOCATION 16K IS UPSTREAM FROM THE ARGONNE SITE AND LOCATION 7H IS DOWNSTREAM FROM THE ARGONNE WASTE-WATER OUTFALL.

** URANIUM CONCENTRATIONS IN UNITS OF MICROGRAMS/L CAN BE OBTAINED BY MULTIPLYING THE CONCENTRATION GIVEN BY 1.48×10^{-9} .

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averages, for the two sampling locations shows the nuclides found in the Creek water that can be attributed to Argonne operations were hydrogen-3, strontium-90, neptunium-237, plutonium-239, americium-241, and occasionally cesium-134, cesium-137, plutonium-238, curium-242 and/or californium-252, and curium-244 and/or californium-249. The percentage of individual samples containing activity attributable to Argonne was 80% for hydrogen-3, 56% for neptunium-237, 60% for plutonium-239, and 90% for americium-241. The concentrations of all these nuclides were low and resulted in very small potential doses. The total downstream concentration, regardless of source, must be used in assessing the hazard of a radionuclide not naturally present. The principal radionuclide added to the Creek by Argonne waste water, in terms of concentration, was hydrogen-3.

The elevated annual average concentrations in the total beta, hydrogen-3, and strontium-90 activities were due to the inadvertent release of these constituents during August and early September. In addition to the above, cesium-134 and cesium-137 were also identified in the release. The maximum concentrations were 6 pCi/L for cesium-134 and 40 pCi/L for cesium-137. Maximum concentrations for all of the above occurred in the samples collected during the second week in August. The Laboratory is currently reviewing its procedures for the control of the disposal of radioactive liquids to prevent future releases.

The hydrogen-3 in the Creek above the site was similar in concentration to levels found away from the Laboratory site and is characteristic of the current ambient level in surface water. During 1986, the hydrogen-3 content of 19 other lakes and streams ranged from $< 100 \times 10^{-9}$ $\mu\text{Ci/mL}$ to 183×10^{-9} $\mu\text{Ci/mL}$ and averaged 120×10^{-9} $\mu\text{Ci/mL}$. After the Marion Brook treatment plant stopped operating, the beta activity decreased by a factor of three in the Creek above the site and this change is assumed to be due to the absence of natural radioactivities in human excretions processed and discharged by the treatment plant.

The total radioactive effluent discharged to the Creek in Argonne waste water can be estimated from the average net concentrations and the

volume of water carried by the Creek. These totals are 31 Ci of hydrogen-3, 0.04 Ci of strontium-90, 0.01 mCi of neptunium-237, 0.02 mCi of plutonium-239, 0.14 mCi of americium-241, and < 0.01 mCi of curium and californium nuclides.

Because Sawmill Creek empties into the Des Plaines River, which in turn flows into the Illinois River, the radioactivity in the two Rivers is important in assessing the contribution of Argonne waste water to the environmental radioactivity. The Des Plaines River was sampled twice a month below, and monthly above, the mouth of Sawmill Creek to determine if the radioactivity in the Creek had any effect on the activity in the River.

Annual summaries of the results obtained for these two locations are given in Table 10. The average nonvolatile alpha, beta, and uranium concentrations in the River were very similar to past averages and remained in the normal range. Results were quite similar above and below the Creek for all radionuclides since the activity in Sawmill Creek was reduced by dilution so that it was not detectable as such in the Des Plaines River. The average nonvolatile alpha and beta activities, 1.2×10^{-9} $\mu\text{Ci/mL}$ and 8.2×10^{-9} $\mu\text{Ci/mL}$, respectively, of 19 off-site surface water samples collected this year (excluding the Des Plaines River) were similar to the levels found in previous years.

The radioactivity in samples of Illinois River water, shown in Table 11, was similar to those found previously at these same locations. No radioactivity originating at Argonne could be detected in the Des Plaines or Illinois Rivers.

A pipe was discovered to be discharging water into the Waterfall Glen Nature Preserve about 200 feet south of the 317 Facility fence, Location 6I/J (see Figure 1). A sample was collected on November 14, 1986, and radiochemical analysis indicated the presence of: $(107 \pm 1) \times 10^{-9}$ $\mu\text{Ci/mL}$ of nonvolatile beta, $(2822 \pm 128) \times 10^{-9}$ $\mu\text{Ci/mL}$ of hydrogen-3, $(31.5 \pm 4.4) \times 10^{-9}$ $\mu\text{Ci/mL}$ of strontium-90, and $(19.6 \pm 2.2) \times 10^{-9}$ $\mu\text{Ci/mL}$ of cesium-137. The source of the water was found to be from the footing drains around the radioactive waste storage vaults within the

TABLE 10
RADIONUCLIDES IN DESPLAINES RIVER WATER, 1986

TYPE OF ACTIVITY	LOCATION *	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/HL)			AVG.	DOSE (MREM)	
			AVG.	MIN.	MAX.		MIN.	MAX.
ALPHA (NONVOLATILE)	A	12	1.5 ± 0.1	0.8	1.9	-	-	-
	B	24	1.7 ± 0.2	0.9	3.4	-	-	-
BETA (NONVOLATILE)	A	12	11 ± 1	6	19	-	-	-
	B	24	12 ± 2	5	21	-	-	-
HYDROGEN-3	A	12	< 113	< 100	223	< 0.0052	< 0.005	0.0103
	B	24	< 114	< 100	170	< 0.0052	< 0.005	0.0078
STRONTIUM-90	A	12	0.31 ± 0.01	< 0.25	0.49	0.03	< 0.02	0.05
	B	24	0.31 ± 0.03	< 0.25	0.44	0.03	< 0.02	0.04
URANIUM (NATURAL) **	A	12	0.9 ± 0.1	0.3	1.3	(0.16)	(0.047)	(0.23)
	B	24	1.0 ± 0.2	0.3	2.1	(0.16)	(0.059)	(0.35)
NEPTUNIUM-237	A	12	-	-	< 0.001	-	-	< 0.00028
	B	12	-	-	< 0.001	-	-	< 0.00028
PLUTONIUM-238	A	12	-	-	< 0.001	-	-	< 0.00028
	B	12	< 0.0010	< 0.001	0.001	0.00029	< 0.00028	0.00039
PLUTONIUM-239	A	12	-	-	< 0.001	-	-	< 0.0003
	B	12	< 0.0011	< 0.001	0.002	< 0.0003	< 0.0003	0.0007
AMERICIUM-241	A	12	-	-	< 0.001	-	-	< 0.0016
	B	12	-	-	< 0.001	-	-	< 0.0016
CURIUM-242 AND/OR CALIFORNIUM-252	A	12	-	-	< 0.001	-	-	< 0.0003
	B	12	-	-	< 0.001	-	-	< 0.0003
CURIUM-244 AND/OR CALIFORNIUM-249	A	12	-	-	< 0.001	-	-	< 0.0017
	B	12	-	-	< 0.001	-	-	< 0.0017

* LOCATION A, NEAR WILLOW SPRINGS, IS UPSTREAM AND LOCATION B, NEAR LEMONT, IS DOWNSTREAM FROM THE MOUTH OF SAWMILL CREEK. SEE FIGURE 2.

** URANIUM CONCENTRATIONS IN UNITS OF MICROGRAMS/L CAN BE OBTAINED BY MULTIPLYING THE CONCENTRATION GIVEN BY 1.48×10^{-9} .

TABLE 11

Radionuclides in Illinois River Water, 1986
(Concentrations in 10^{-9} $\mu\text{Ci/mL}$)

Date Collected	Location	Alpha*	Beta*	Hydrogen-3	Uranium** (natural)	Plutonium-239
June 5	McKinley Woods State Park	1.5 \pm 0.3	8.8 \pm 0.3	< 100	0.7 \pm 0.1	< 0.001
June 5	Below Dresden Power Station	0.9 \pm 0.2	7.4 \pm 0.3	< 100	0.8 \pm 0.1	< 0.001
June 5	Morris	1.2 \pm 0.2	7.6 \pm 0.3	< 100	0.7 \pm 0.1	-
June 5	Starved Rock State Park	1.2 \pm 0.2	5.9 \pm 0.3	183 \pm 97	0.8 \pm 0.1	-
October 15	McKinley Woods State Park	0.7 \pm 0.2	7.9 \pm 0.3	184 \pm 76	0.6 \pm 0.1	< 0.001
October 15	Below Dresden Power Station	1.1 \pm 0.2	4.5 \pm 0.3	183 \pm 76	1.0 \pm 0.1	< 0.001
October 15	Morris	1.1 \pm 0.3	6.9 \pm 0.4	218 \pm 76	1.1 \pm 0.1	-
October 15	Starved Rock State Park	1.2 \pm 0.2	5.9 \pm 0.3	126 \pm 75	0.9 \pm 0.1	-

* Nonvolatile activity.

** Uranium concentrations in units of $\mu\text{g/L}$ can be obtained by multiplying the concentration by 1.48×10^9 .

317 Area. The water drained south of the site, but did not reach the Des Plaines River. The pipe was plugged and a complete characterization of the area is planned for 1987.

3. Ground Water

The Laboratory domestic water is provided by four wells which are described in Section I.E. and the locations are shown in Figure 1. Samples from each well were collected quarterly at the well head and analyzed for several types of radioactivity. The results are in Table 12. In addition to the well water samples, one tap water sample was collected and the results are also in Table 12.

Since the Laboratory is a "non-community water system",¹² the EPA standards for this type of system apply. For the nuclides measured in Table 12, the EPA limits are:

Gross alpha particle activity	15 pCi/L (10^{-9} μ Ci/mL)
Gross beta particle activity	50 pCi/L (10^{-9} μ Ci/mL)
Hydrogen-3	2×10^4 pCi/L (10^{-9} μ Ci/mL)
Strontium-90	8 pCi/L (10^{-9} μ Ci/mL)
Radium-226	5 pCi/L (10^{-9} μ Ci/mL)

The uranium results would be covered by the gross alpha standard. Inspection of Table 12 indicates that all measurements are well within the EPA drinking water standards. This program is being conducted to demonstrate the Laboratory's compliance with the EPA drinking water regulations.

Wells 1 and 2 had measurable levels of hydrogen-3 at various times during the year, although the average concentration was about 1% of the EPA Standard. It is speculated that the source of the hydrogen-3 was from liquid wastes that were placed in holding ponds in the sewage treatment area (Location 10M in Figure 1) in the 1950's. The tritiated water migrated down through the soil to the dolomite, and was drawn into the wells. Well 1, which is about 200 m north of the treatment area, had higher hydrogen-3 concentrations than Well 2, which is about 300 m from the treatment

TABLE 12

Radioactivity in ANL Domestic Wells, 1986
(Concentrations in 10^{-9} $\mu\text{Ci/mL}$)

Type of Activity	Location	No. of Samples	Avg.	Min.	Max.
Alpha (nonvolatile)	Well #1	3	3.3	1.8	4.5
	Well #2	4	4.3	1.2	6.6
	Well #3	4	2.8	1.1	4.3
	Well #4	4	2.6	1.7	3.0
	Tap	1	-	-	0.4
Beta (nonvolatile)	Well #1	3	6.2	4.2	7.6
	Well #2	4	8.0	4.9	9.7
	Well #3	4	6.7	5.3	8.1
	Well #4	4	6.4	5.9	7.3
	Tap	1	-	-	5.2
Hydrogen-3	Well #1	3	155	< 100	239
	Well #2	4	218	< 100	478
	Well #3	4	117	< 100	169
	Well #4	4	114	< 100	158
	Tap	1	-	-	147
Strontium-90	Well #1	1	-	-	< 0.25
	Well #2	1	-	-	< 0.25
	Well #3	1	-	-	< 0.25
	Well #4	1	-	-	< 0.25
	Tap	1	-	-	< 0.25
Radium-226	Well #1	1	-	-	1.26
	Well #2	1	-	-	1.66
	Well #3	1	-	-	0.69
	Well #4	1	-	-	0.51
	Tap	1	-	-	< 0.10
Uranium (natural)	Well #1	1	-	-	0.44
	Well #2	1	-	-	0.33
	Well #3	1	-	-	0.41
	Well #4	1	-	-	0.22
	Tap	1	-	-	0.06

area. Although the normal subsurface water flow gradient is in the south direction, the cone of depression created by the pumping on these wells would overpower the normal pattern. The holding ponds have not been used for a number of years. One hydrogen-3 result from Well 3 and one from Well 4 were very slightly above the detection limit, but these are considered to be within the normal fluctuation range for the measurement of hydrogen-3 in water.

In the 1982 monitoring report,¹³ an unusual occurrence was reported which consisted of the loss of about 7.5 megaliters (2 million gallons) of ponded water containing about 26 mCi of tritiated water from the Argonne Advanced Research Reactor (A²R²) excavation (Location 10G). Quarterly monitoring of wells and a seep likely to be in the subsurface flow path of this water continued during 1986 and the results are summarized in Table 13. The concentrations of tritiated water were in the normal range and none was detected that could be attributed to this release. In 1982, elevated concentrations, up to 1200×10^{-9} $\mu\text{Ci/mL}$, were measured in Well 9 from this release. The concentration in the tritiated water that remains in the A²R² excavation continued to decrease from the 2.2×10^{-6} $\mu\text{Ci/mL}$ in 1982 to 0.6×10^{-6} $\mu\text{Ci/mL}$ in 1986. The excavation is being filled with concrete, asphalt, and soil as these materials become available. Well 9 and Well 10 are used to measure water levels, and are not a source of drinking water.

During the fall of the year, three additional wells were added and three wells replaced at the Laboratory sanitary landfill, see Section III. B.2 and Figure 7. Samples collected from the sanitary landfill monitoring wells on October 22, 1986, and December 23, 1986, for chemical analysis were also analyzed for tritiated water. The results are in Table 14. Although the disposal of radioactive material is prohibited in the sanitary landfill, very low concentrations of tritiated water were detected, probably due to their inadvertent disposal with Laboratory trash. However, the presence of the tritiated water provides information on the subsurface water flow pathway in the sanitary landfill area. The data indicated that the principal direction of subsurface water flow is south-southeast, with a

TABLE 13

Tritium Content of Water Near
 $A^{235}R^{235}$ Excavation, 1986
 (Concentrations in 10^{-9} $\mu\text{Ci/mL}$)

Sample	Location*	No. of Samples	Avg.	Min.	Max.
Well 9	9F	4	105	< 100	121
Well 10	11J	4	-	-	< 100
Well D181	8F	4	-	-	< 100
Well D185	8E	3	115	< 100	145
Seep	4EF	4	122	< 100	186
$A^{235}R^{235}$	10G	4	594	546	661

* See Figure 1.

TABLE 14

Hydrogen-3 Content of Water From Sanitary Landfill Wells
 (Concentrations in 10^{-9} $\mu\text{Ci/mL}$)

Location	Date Collected	
	October 22, 1986	December 23, 1986
Well 1-2	234 \pm 93	267 \pm 111
Well 2-2	201 \pm 92	323 \pm 112
Well 3	< 100	< 100
Well 4-2	< 100	163 \pm 109
Well 5	486 \pm 97	592 \pm 116
Well 6	557 \pm 98	517 \pm 115
Well 7a	759 \pm 102	817 \pm 120
Well 8	181 \pm 92	168 \pm 109
Well 9	950 \pm 104	711 \pm 118
Well 10	< 100	823 \pm 108

small component to the northwest. This is consistent with estimates based on water level measurements and general flow patterns in the area.

4. Soil, Grass, and Bottom Sediment

The radioactive content of soil, grass, and bottom sediment was measured at the site perimeter and off the site. The reason for the off-site sampling was to measure deposition for comparison with perimeter samples, and with results obtained by other organizations for samples collected at large distances from nuclear installations. Such comparisons are useful in determining if the soil activity near Argonne is normal. For this purpose, the American Society for Testing and Materials (ASTM) site selection criteria, sample collection, and sample preparation techniques were used.¹⁴ Sites were selected in several directions and at various distances from the Laboratory. Each site was selected on the basis that the soil appeared, or was known to have been, undisturbed for a number of years. Attempts were made to select open, level, grassy areas that were mowed at reasonable intervals. Public parks were selected when available.

Each soil sample consisted of ten cores totaling 864 cm^2 in area by 5 cm deep. Through 1976, samples had been collected down to 30 cm to measure total deposition and as a result of five years of sample collection at this depth, the total deposition in the Argonne environment has been established. By reducing the sampling depth to 5 cm, the analysis will be more sensitive to changes in current deposition. The grass samples were obtained by collecting the grass from a 1 m^2 area in the immediate vicinity of a soil sample. A grab sample technique was used to obtain bottom sediment. After drying, grinding, and mixing, 100 g portions of soil, bottom sediment, and grass were analyzed by the same methods described in Section III.A.1 for air-filter residues. The plutonium and americium were separated from the same 100 gram aliquot of soil. Results are given in terms of the oven-dried (110°C) weight.

The results for the gamma-ray emitting nuclides in soil are presented in Table 15. Intermediate half-life fission products reported previously

TABLE 15
 Gamma-Ray Emitting Radionuclides in Soil, 1986
 (Concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location	Potassium-40	Cesium-137	Radium-226	Thorium-228	Thorium-232
	<u>Perimeter</u> *					
June 5	5D	16.68 \pm 0.57	0.70 \pm 0.03	1.06 \pm 0.06	0.91 \pm 0.03	0.71 \pm 0.08
June 5	8N	18.96 \pm 0.61	0.34 \pm 0.02	1.23 \pm 0.06	0.87 \pm 0.03	0.81 \pm 0.08
June 5	12-0	14.69 \pm 0.61	0.97 \pm 0.04	1.14 \pm 0.06	0.91 \pm 0.04	0.81 \pm 0.09
June 5	14L	15.79 \pm 0.56	0.60 \pm 0.03	1.14 \pm 0.06	0.88 \pm 0.03	0.83 \pm 0.08
June 5	12C	15.51 \pm 0.55	0.84 \pm 0.03	1.32 \pm 0.06	0.86 \pm 0.03	0.88 \pm 0.09
June 24	7J	17.51 \pm 0.59	0.96 \pm 0.04	1.09 \pm 0.06	0.95 \pm 0.03	0.83 \pm 0.08
June 24	7J	18.82 \pm 0.61	2.15 \pm 0.05	1.06 \pm 0.06	0.94 \pm 0.03	0.86 \pm 0.08
June 24	7J	17.03 \pm 0.57	0.76 \pm 0.03	1.14 \pm 0.06	0.94 \pm 0.03	0.83 \pm 0.08
October 27	10E	18.11 \pm 0.77	0.84 \pm 0.04	1.40 \pm 0.07	1.02 \pm 0.04	0.83 \pm 0.09
October 27	9N	16.62 \pm 0.59	0.85 \pm 0.04	1.25 \pm 0.06	1.01 \pm 0.04	0.78 \pm 0.08
October 27	10N	14.75 \pm 0.55	0.45 \pm 0.03	1.07 \pm 0.06	0.68 \pm 0.03	0.59 \pm 0.07
October 28	6J	22.37 \pm 0.78	0.76 \pm 0.04	1.43 \pm 0.07	1.15 \pm 0.04	0.89 \pm 0.09
October 28	14I	17.58 \pm 0.60	0.85 \pm 0.04	1.31 \pm 0.06	1.04 \pm 0.04	0.90 \pm 0.09
	Average	17.26 \pm 1.15	0.85 \pm 0.24	1.20 \pm 0.07	0.94 \pm 0.08	0.81 \pm 0.05
	<u>Off-Site</u>					
June 3	West Chicago, IL	14.55 \pm 0.67	0.44 \pm 0.03	1.47 \pm 0.08	1.07 \pm 0.04	1.02 \pm 0.11
June 3	Naperville, IL	13.79 \pm 0.66	0.61 \pm 0.04	1.49 \pm 0.08	2.93 \pm 0.07	2.57 \pm 0.15
June 3	Lemont, IL	16.55 \pm 0.58	1.08 \pm 0.04	1.51 \pm 0.06	0.79 \pm 0.03	0.69 \pm 0.08
June 5	Channahon, IL	15.51 \pm 0.56	0.81 \pm 0.03	1.13 \pm 0.06	0.95 \pm 0.03	0.93 \pm 0.08
June 5	Starved Rock State Park, IL	15.31 \pm 0.55	0.34 \pm 0.02	1.98 \pm 0.07	0.89 \pm 0.03	0.83 \pm 0.08
October 15	McKinley Woods State Park, IL	18.52 \pm 0.75	1.81 \pm 0.05	1.41 \pm 0.07	0.86 \pm 0.04	0.79 \pm 0.09
October 15	Morris, IL	16.18 \pm 0.72	0.32 \pm 0.03	1.62 \pm 0.07	1.06 \pm 0.04	0.89 \pm 0.09
October 15	Dresden Lock & Dam, IL	22.06 \pm 0.65	0.98 \pm 0.04	1.46 \pm 0.06	1.08 \pm 0.04	0.88 \pm 0.08
October 16	Western Springs, IL	20.21 \pm 0.77	0.65 \pm 0.03	1.89 \pm 0.07	1.06 \pm 0.04	0.83 \pm 0.09
October 16	Brookfield, IL	17.93 \pm 0.60	1.18 \pm 0.04	1.48 \pm 0.06	0.93 \pm 0.04	0.71 \pm 0.08
	Average	17.06 \pm 1.65	0.82 \pm 0.29	1.54 \pm 0.15	1.16 \pm 0.40	1.04 \pm 0.35

*The perimeter locations are given in terms of the grid coordinates in Figure 1.

have decayed to below their detection limits and no evidence of Chernobyl fallout is apparent. The cesium-137 levels are similar to those found over the past several years, and represent an accumulation from nuclear tests over a period of many years. The annual average concentrations for the perimeter and off-site samples are similar. The plutonium and americium concentrations are given in Table 16. Except for the samples collected June 24, 1986, from Location 7J, the ranges and concentrations of plutonium and americium in soil are similar at both perimeter and off-site locations. A sample collected from Location 7J in 1985 gave plutonium and americium concentrations that were twice the ambient levels. The results of the analyses of the three samples collected this year, at the south perimeter of the 317 Area, confirm the previous measurement. The highest plutonium-239 concentration, 80×10^{-9} $\mu\text{Ci/g}$, was collected in line with the south-east corner of the 317 Area. This radioactivity appears to be the result of previous operations in this area. For fallout americium-241 in soil, about 10% is due to direct deposition, while about 90% is from the decay of the previously deposited plutonium-241.¹⁵ The measured deposition of americium-241 and the americium-241/plutonium-239 ratio is consistent with reported values.¹⁵

The results of radionuclide concentrations measured in grass are given in Table 17. The annual averages and concentration ranges were similar at the perimeter and off-site locations as well as similar to previous years, indicating no contribution from ANL operations. In terms of deposition, the plutonium-239 concentration was a factor of about 10^4 less in the grass than in the soil from the same location. Elevated cesium-137 concentrations in the June sampling were probably due to Chernobyl fallout, which had been washed into the soil by the time of the fall sampling. The concentration of cesium-137 added to soil by the Chernobyl accident was small compared to that already present from previous atmospheric nuclear tests.

Results of analyses of bottom sediment samples for gamma-ray emitters and transuranics are given in Table 18. The annual off-site averages are in the same range found in off-site samples collected in previous years. The sample with the highest plutonium-239 concentration, Lower Freund Pond Outlet, Location 11/12K, has been observed to have higher concentrations in

TABLE 16

Transuranics in Soil, 1986

Date Collected	Location	Plutonium-238		Plutonium-239		$^{238}\text{Pu}/^{239}\text{Pu}$	Americium-241		$^{241}\text{Am}/^{239}\text{Pu}$
		10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$		10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	
	<u>Perimeter*</u>								
June 5	5D	0.6 ± 0.2	0.033 ± 0.008	13.4 ± 0.7	0.717 ± 0.037	0.045	3.2 ± 1.2	0.169 ± 0.067	0.24
June 5	8N	0.4 ± 0.1	0.031 ± 0.010	5.7 ± 0.4	0.413 ± 0.032	0.076	0.5 ± 0.9	0.034 ± 0.063	0.08
June 5	12-O	0.8 ± 0.2	0.039 ± 0.010	20.3 ± 1.0	0.949 ± 0.045	0.041	6.9 ± 1.9	0.322 ± 0.087	0.34
June 5	14L	0.6 ± 0.2	0.031 ± 0.009	12.7 ± 0.7	0.674 ± 0.039	0.046	3.2 ± 1.1	0.172 ± 0.059	0.26
June 5	12C	0.6 ± 0.2	0.031 ± 0.009	16.7 ± 0.8	0.888 ± 0.045	0.035	3.9 ± 1.8	0.205 ± 0.097	0.23
June 24	7J	0.9 ± 0.2	0.040 ± 0.009	56.0 ± 1.5	2.555 ± 0.067	0.016	4.9 ± 1.4	0.223 ± 0.063	0.09
June 24	7J	2.2 ± 0.3	0.096 ± 0.013	80.4 ± 1.8	3.509 ± 0.078	0.027	14.2 ± 2.0	0.617 ± 0.089	0.18
June 24	7J	0.8 ± 0.2	0.047 ± 0.015	19.7 ± 1.1	1.220 ± 0.067	0.038	3.4 ± 1.0	0.212 ± 0.061	0.17
October 27	10E	0.7 ± 0.2	0.038 ± 0.012	20.1 ± 1.0	1.035 ± 0.052	0.037	-	-	-
October 27	9N	0.6 ± 0.2	0.032 ± 0.011	19.2 ± 1.0	1.014 ± 0.052	0.032	-	-	-
October 27	10N	0.6 ± 0.2	0.031 ± 0.011	11.1 ± 0.8	0.534 ± 0.037	0.058	-	-	-
October 28	6J	0.6 ± 0.2	0.035 ± 0.013	17.5 ± 1.0	1.006 ± 0.057	0.035	-	-	-
October 28	14I	0.7 ± 0.2	0.033 ± 0.010	18.4 ± 0.9	0.909 ± 0.045	0.036	-	-	-
	Average	0.8 ± 0.3	0.040 ± 0.011	23.9 ± 12.9	1.186 ± 0.542	0.040	5.0 ± 3.6	0.244 ± 0.148	0.20
	<u>Off-Site</u>								
June 3	West Chicago, IL	0.4 ± 0.1	0.016 ± 0.005	10.6 ± 0.6	0.415 ± 0.024	0.038	2.7 ± 0.8	0.107 ± 0.032	0.26
June 3	Naperville, IL	1.1 ± 0.2	0.057 ± 0.011	10.9 ± 0.6	0.564 ± 0.032	0.101	4.6 ± 0.6	0.237 ± 0.030	0.42
June 3	Lemont, IL	0.8 ± 0.2	0.032 ± 0.007	19.1 ± 0.9	0.724 ± 0.033	0.044	5.4 ± 0.3	0.204 ± 0.011	0.28
June 5	Channahon, IL	0.7 ± 0.2	0.027 ± 0.007	18.6 ± 0.8	0.758 ± 0.034	0.036	4.4 ± 1.2	0.386 ± 0.050	0.24
June 5	Starved Rock State Park, IL	0.3 ± 0.2	0.013 ± 0.007	3.3 ± 0.4	0.156 ± 0.016	0.053	1.1 ± 1.2	0.052 ± 0.056	0.34
October 15	McKinley Woods State Park, IL	1.2 ± 0.4	0.044 ± 0.014	35.3 ± 1.7	1.329 ± 0.064	0.033	-	-	-
October 15	Morris, IL	0.2 ± 0.2	0.009 ± 0.009	7.4 ± 0.7	0.432 ± 0.039	0.021	-	-	-
October 15	Dresden Lock and Dam, IL	0.8 ± 0.2	0.029 ± 0.009	22.0 ± 1.1	0.844 ± 0.042	0.034	-	-	-
October 16	Western Springs, IL	0.9 ± 0.3	0.052 ± 0.016	17.0 ± 1.0	1.011 ± 0.061	0.051	-	-	-
October 16	Brookfield, IL	1.0 ± 0.2	0.057 ± 0.013	23.0 ± 1.0	1.305 ± 0.057	0.044	-	-	-
	Average	0.7 ± 0.2	0.034 ± 0.010	16.7 ± 5.8	0.754 ± 0.242	0.046	3.6 ± 2.2	0.197 ± 0.166	0.31

*The perimeter locations are given in terms of the grid coordinates in Figure 1.

TABLE 17

Radionuclides in Grass, 1986

Date Collected	Location	(Concentrations in 10^{-9} $\mu\text{Ci/g}$)		(10^{-6} $\mu\text{Ci/m}^2$)
		Cesium-137	Plutonium-239	Deposition of Plutonium-239
	<u>Perimeter</u> *			
June 5	5D	204 \pm 19	< 0.1	< 0.01
June 5	8N	291 \pm 21	0.1 \pm 0.1	0.03 \pm 0.01
June 5	12-0	373 \pm 24	< 0.1	< 0.01
June 5	12L	155 \pm 23	0.4 \pm 0.1	0.16 \pm 0.04
June 5	12C	219 \pm 20	0.1 \pm 0.1	0.02 \pm 0.01
October 27	10E	18 \pm 20	0.3 \pm 0.1	0.04 \pm 0.02
October 27	9N	36 \pm 21	0.1 \pm 0.1	0.04 \pm 0.02
October 27	10N	9 \pm 18	< 0.1	< 0.01
October 28	6J	33 \pm 13	0.1 \pm 0.1	0.01 \pm 0.01
October 28	14I	74 \pm 22	< 0.1	< 0.01
	Average	141 \pm 81	0.1 \pm 0.1	0.03 \pm 0.02
	<u>Off-Site</u>			
June 3	West Chicago, IL	231 \pm 21	< 0.1	< 0.01
June 3	Naperville, IL	91 \pm 16	< 0.1	< 0.01
June 3	Lemont, IL	73 \pm 16	< 0.1	< 0.01
June 5	Channahon, IL	291 \pm 21	< 0.1	< 0.01
June 5	Starved Rock State Park, IL	291 \pm 26	0.1 \pm 0.1	0.02 \pm 0.01
October 15	McKinley Woods State Park, IL	43 \pm 19	0.7 \pm 0.1	0.06 \pm 0.01
October 15	Morris, IL	48 \pm 21	0.5 \pm 0.2	0.04 \pm 0.02
October 15	Dresden Lock and Dam, IL	11 \pm 12	0.5 \pm 0.1	0.04 \pm 0.01
October 16	Western Springs, IL	26 \pm 19	0.2 \pm 0.1	0.02 \pm 0.01
October 16	Brookfield, IL	86 \pm 28	0.9 \pm 0.2	0.06 \pm 0.01
	Average	119 \pm 69	0.3 \pm 0.2	0.03 \pm 0.02

*The perimeter locations are given in terms of the grid coordinates in Figure 1.

past samplings. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material. A set of samples was collected on June 24, 1986, from the Sawmill Creek bed, above, at, and at several locations below the point at which the Laboratory discharges its treated waste water (Location 7M in Figure 1). The results are listed in Table 18 and indicate that the results of the sample above the 7M outfall are similar to the off-site samples. The plutonium and americium concentrations are the highest at the outfall and decrease rapidly with distance, indicating their origin is in Argonne waste water. Similar sets of samples were collected on June 17, 1985, July 27, 1984, August 11, 1983, September 15, 1982, and September 24, 1980. Comparison of plutonium concentrations indicates that the 1982 results were an order of magnitude higher at the outfall, but similar farther downstream, while the 1980, 1983, 1984, and 1985 results were more like the 1986 set at the same locations. The changes in concentrations of the various nuclides with time indicates the dynamic nature of the sediment material in this area.

5. Milk

Raw milk was collected monthly from a local dairy farm south of Lemont and analyzed for several radionuclides. The water was separated from the milk by low-temperature vacuum evaporation and the hydrogen-3 determined by liquid scintillation spectrometry. The strontium-90 was analyzed by the same method used for water and with the same detection limit. The results are given in Table 19. The average strontium-90 concentration was similar to the 1985 results. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations.

The concentrations given in Table 19 may be compared to the EPA drinking water limits. The consumption of one liter of milk per day would result in an average dose of 0.6 mrem/y for strontium-90 and < 0.25 mrem/y for hydrogen-3.

TABLE 18

Radionuclides in Bottom Sediment, 1986

Date Collected	Location	Concentrations in 10^{-6} $\mu\text{Ci/g}$					Concentrations in 10^{-9} $\mu\text{Ci/g}$			
		Potassium-40	Cesium-137	Radium-226	Thorium-228	Thorium-232	Plutonium-238	Plutonium-239	Americium-241	
<u>Perimeter</u> ^a										
June 19	Building 205 Pond - 11C	14.99 \pm 0.56	0.37 \pm 0.02	0.86 \pm 0.05	0.67 \pm 0.03	0.62 \pm 0.07	0.6 \pm 0.2	7.8 \pm 0.5	3.3 \pm 1.2	
June 20	Upper Freund Pond - 11/12K	20.27 \pm 0.79	0.32 \pm 0.03	1.46 \pm 0.07	0.90 \pm 0.04	0.76 \pm 0.09	0.8 \pm 0.3	12.3 \pm 1.4	3.2 \pm 0.8	
June 20	Lower Freund Pond Inlet - 12K	21.79 \pm 0.75	0.38 \pm 0.03	1.64 \pm 0.07	1.02 \pm 0.04	0.93 \pm 0.09	0.6 \pm 0.2	20.5 \pm 1.2	4.1 \pm 1.5	
June 20	Lower Freund Pond Outlet - 11/12K	19.45 \pm 0.85	0.65 \pm 0.04	1.26 \pm 0.08	0.87 \pm 0.04	0.68 \pm 0.10	1.2 \pm 0.2	57.6 \pm 2.6	7.0 \pm 1.4	
June 24	Sawmill Creek at Des Plaines River	12.44 \pm 0.72	0.99 \pm 0.04	1.05 \pm 0.06	0.63 \pm 0.03	0.58 \pm 0.08	1.5 \pm 0.3	19.1 \pm 1.3	6.4 \pm 0.4	
June 24	Sawmill Creek - 100 m Below Outfall	8.55 \pm 0.43	0.34 \pm 0.02	0.84 \pm 0.05	0.38 \pm 0.03	0.38 \pm 0.06	0.5 \pm 0.2	10.8 \pm 0.7	3.5 \pm 0.3	
June 24	Sawmill Creek - 50 m Below Outfall	7.60 \pm 0.41	0.16 \pm 0.02	0.73 \pm 0.05	0.35 \pm 0.02	0.41 \pm 0.06	0.4 \pm 0.2	10.1 \pm 0.7	1.5 \pm 0.2	
June 24	Sawmill Creek - Outfall	6.24 \pm 0.40	0.26 \pm 0.02	0.71 \pm 0.05	0.29 \pm 0.02	0.41 \pm 0.07	0.8 \pm 0.2	10.4 \pm 0.8	1.3 \pm 0.2	
June 24	Sawmill Creek - 50 m Above Outfall	13.20 \pm 0.71	0.41 \pm 0.03	1.34 \pm 0.07	0.68 \pm 0.03	0.76 \pm 0.09	0.3 \pm 0.1	8.3 \pm 0.6	2.5 \pm 0.2	
June 25	Stream at 7/8K Perimeter Fence	15.47 \pm 0.53	0.16 \pm 0.02	1.09 \pm 0.06	0.75 \pm 0.03	0.60 \pm 0.07	0.1 \pm 0.1	3.2 \pm 0.4	2.1 \pm 0.8	
November 11	Stream at 7/8K Perimeter Fence	15.01 \pm 0.55	0.08 \pm 0.02	1.38 \pm 0.05	0.76 \pm 0.03	0.66 \pm 0.07	< 0.1	1.8 \pm 0.4	-	
<u>Off-Site</u>										
June 3	DuPage River, West Chicago, IL	16.12 \pm 0.72	0.27 \pm 0.03	1.98 \pm 0.07	1.24 \pm 0.04	1.20 \pm 0.10	0.2 \pm 0.1	4.9 \pm 0.3	1.6 \pm 0.6	
June 3	DuPage River, Naperville, IL	7.94 \pm 0.41	0.18 \pm 0.02	0.95 \pm 0.05	1.33 \pm 0.04	1.17 \pm 0.09	0.3 \pm 0.1	3.5 \pm 0.3	< 0.1	
June 3	Long Run Creek, Lemont, IL	20.68 \pm 0.77	0.19 \pm 0.02	2.22 \pm 0.08	1.18 \pm 0.04	0.95 \pm 0.09	0.2 \pm 0.1	4.7 \pm 0.3	2.2 \pm 0.5	
June 5	DuPage River, Channahon, IL	9.23 \pm 0.44	0.05 \pm 0.02	0.92 \pm 0.05	1.02 \pm 0.03	0.78 \pm 0.08	< 0.1	1.0 \pm 0.2	1.3 \pm 0.3	
June 5	Illinois River, Starved Rock State Park, IL	5.77 \pm 0.37	0.09 \pm 0.02	0.80 \pm 0.05	0.40 \pm 0.03	0.40 \pm 0.07	0.1 \pm 0.1	0.9 \pm 0.1	0.4 \pm 0.1	
June 27	Pond, West Side of Lemont Rd, @ 75th St.	16.36 \pm 0.77	0.79 \pm 0.04	1.28 \pm 0.07	0.88 \pm 0.04	0.73 \pm 0.09	0.6 \pm 0.2	19.2 \pm 0.9	5.2 \pm 1.0	
October 15	Illinois River, McKinley Woods, IL	9.58 \pm 0.45	0.03 \pm 0.01	0.47 \pm 0.04	0.34 \pm 0.02	0.36 \pm 0.06	< 0.1	0.5 \pm 0.2	-	
October 15	Illinois River, Morris, IL	12.89 \pm 0.53	0.10 \pm 0.02	0.67 \pm 0.05	0.45 \pm 0.03	0.41 \pm 0.07	< 0.1	1.1 \pm 0.2	0.4 \pm 0.2	
October 15	Illinois River, Dresden, IL	15.86 \pm 0.56	0.29 \pm 0.02	0.98 \pm 0.05	0.70 \pm 0.03	0.66 \pm 0.08	< 0.1	2.0 \pm 0.3	1.3 \pm 0.6	
October 16	Des Plaines River, Brookfield, IL	17.17 \pm 0.58	0.47 \pm 0.03	1.31 \pm 0.06	0.94 \pm 0.04	0.90 \pm 0.09	0.2 \pm 0.1	5.4 \pm 0.5	3.0 \pm 0.8	
October 16	Salt Creek, Western Springs, IL	11.31 \pm 0.51	0.10 \pm 0.02	1.98 \pm 0.07	0.80 \pm 0.03	0.88 \pm 0.09	< 0.1	0.9 \pm 0.2	0.7 \pm 0.5	
	Average	12.99 \pm 2.78	0.23 \pm 0.14	1.23 \pm 0.35	0.84 \pm 0.21	0.77 \pm 0.18	0.2 \pm 0.1	4.0 \pm 3.2	1.6 \pm 1.0	

^aThe perimeter locations are given in terms of the grid coordinates in Figure 1.

TABLE 19
 Radionuclides in Milk, 1986
 (Concentrations in 10^{-9} $\mu\text{Ci/mL}$)

Date Collected	Hydrogen-3	Strontium-90
January 8	< 100	2.3 \pm 0.1
February 12	< 100	2.0 \pm 0.3
March 12	< 100	2.1 \pm 0.2
April 4	< 100	2.6 \pm 0.3
May 7	< 100	2.5 \pm 0.3
June 5	< 100	3.9 \pm 0.5
July 2	< 100	3.7 \pm 0.3
August 13	< 100	3.0 \pm 1.1
September 3	< 100	2.9 \pm 0.5
October 1	< 100	3.4 \pm 1.6
November 5	< 100	2.6 \pm 0.4
December 3	< 100	2.6 \pm 0.2
Average	< 100	2.8 \pm 0.3

6. External Penetrating Radiation

Measurements were made with calcium fluoride and lithium fluoride thermoluminescent dosimeter (TLD) chips. Each measurement was the average of four chips exposed in the same packet. All calcium fluoride packets were shielded with 1.6 mm (1/16 in) copper foil to reduce or eliminate the beta and low-energy X-ray components. The response of the chips was determined with a U. S. National Bureau of Standards (NBS) standard radium-226 source, and the results were calculated in terms of air dose. Dosimeters were exposed at a number of locations at the site boundary and on the site. Readings were also taken at five off-site locations for comparison purposes. These locations are shown in Figure 2.

The results are summarized in Tables 20 and 21, and the site boundary and on-site readings are also shown in Figure 5. Measurements were made

for the four successive exposure periods shown in the tables and the results were calculated in terms of annual dose for ease in comparing measurements made for different elapsed times. The uncertainty given in the tables for an average is the 95% confidence limit calculated from the standard deviation of the average.

TABLE 20

Environmental Penetrating Radiation at Off-Site Locations, 1986

Location	Dose Rate (mrem/year)				Average
	1/9-4/10	Period of Measurement		10/14-1/15	
		4/10-7/15	7/15-10/14		
Lemont	66	82	82	83	78 \pm 8
Lombard	67	86	81	83	79 \pm 8
Oak Brook	65	98	84	86	83 \pm 13
Oak Lawn	61	75	71	75	70 \pm 7
Woodridge	63	81	82	80	76 \pm 9
Average	64 \pm 2	84 \pm 9	80 \pm 5	81 \pm 4	78 \pm 5

The off-site results averaged 78 ± 5 mrem/y and are similar to last year's off-site average of 76 ± 5 mrem/y. If the off-site locations are an accurate sample of the radiation background in the area, then annual averages at the site in the range of 78 ± 5 mrem/y are the ambient or normal levels with a 95% probability. To compare boundary results for individual sampling periods, the standard deviation of the 20 individual off-site results is useful. This value is 9.4 mrem/y, so that individual results in the range of 78 ± 19 mrem/y may be considered normal with a 95% probability, unless there are known reasons to the contrary.

At two site boundary locations, 7I (south) and 14I (north), the dose rates were consistently above the normal range. At 7I this was due to radiation from a Radioactive Waste Storage Facility (317) in the northern half of grid 7I. Waste is packaged and temporarily kept in this area prior to

TABLE 21

Environmental Penetrating Radiation at ANL, 1986

Location*	Dose Rate (mrem/year)				Average
	Period of Measurement				
	1/9-4/10	4/10-7/15	7/15-10/14	10/14-1/15	
14L - Boundary	56	71	70	70	67 ± 8
14I - Boundary	93	105	105	105	102 ± 6
14G - Boundary	69	87	83	83	80 ± 8
9/10EF - Boundary	63	79	80	80	76 ± 8
8H - Boundary	66	80	82	78	76 ± 7
8H - Boundary, Center, St. Patrick's Cemetery	76	85	85	87	83 ± 5
7I - Boundary	424	413	389	549	444 ± 72
6I - 200 m N of Quarry Road	65	83	86	79	78 ± 9
9H - 50 m SE of CP-5	512	462	385	385	436 ± 62
8H - 65 m S of Building 316	92	80	79	76	82 ± 7
8H - 200 m NW of Waste Storage Area (Heliport)	93	88	85	92	90 ± 4
7I - Center, Waste Storage Area Facility 317	4080	4400	3610	6340	4610 ± 1200
10/11K - Lodging Facilities	56	71	71	71	67 ± 7
9I - 65 m NE of Building 350, 230 m NE of Building 316	63	106	75	70	78 ± 19

*See Figure 1.

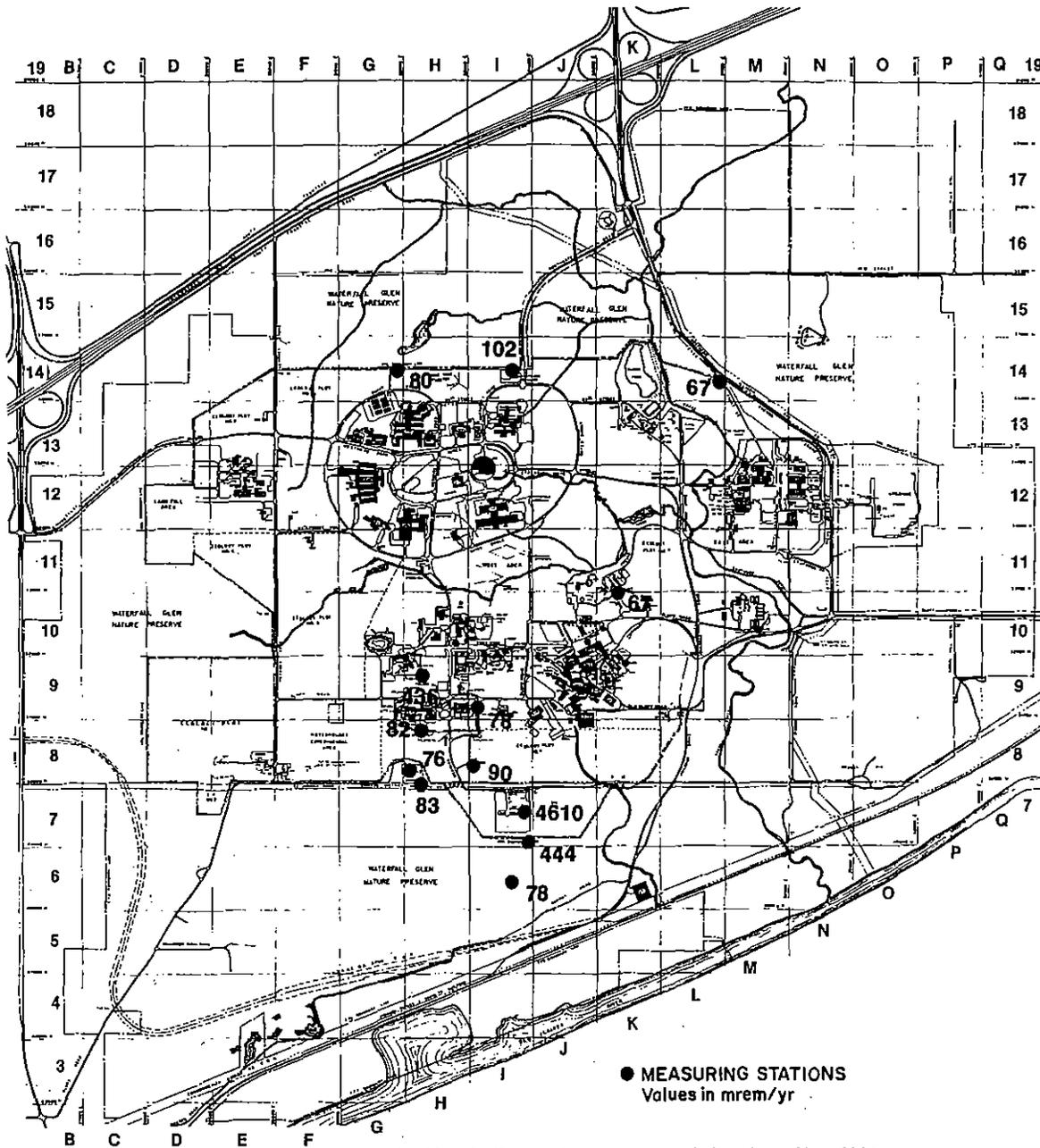


Fig. 5. Penetrating Radiation Measurements at the ANL Site, 1986

removal for permanent storage elsewhere. The net above-normal dose at this location was about 366 mrem/y, about one-half the 1985 average. In previous years, this value has ranged from 865 mrem/y in 1985 to 114 mrem/y in 1977. About 300 m (0.2 mi) south of the fence in grid 6I, the measured dose dropped to 78 ± 9 mrem/y, within the normal range. The above-normal dose at the 8H/I Location, 200 m NW of the Waste Storage Area, about 12 mrem/y, is also due to the Storage Area, as discussed in the 1982 report.¹³ Elevated levels at Location 9H are due to activated equipment from the CP-5 facility stored in the yard next to the building. At Location 14I, at the north boundary, the dose rate was 24 mrem/y above normal, about the same as in 1985. This dose is due to the use of cobalt-60 irradiation sources in Building 202.

The dose in the south portion of grid 8H is of interest. This area includes St. Patrick's Cemetery, which was in use before Argonne was constructed and is open to visitors. In 1986, as in previous years, this dose is estimated to be 5 to 10 mrem/y above the off-site average. Possible explanations are that the 8H dose rates are natural since the differences between the off-site and 8H averages are not statistically significant at the 95% confidence level or that the monument stones in the Cemetery produce the elevated rates because above normal dose rates were obtained from one of the large red granite stones, and granite is known to contain above average levels of natural thorium and its decay products.

7. Potential Radiation Dose Estimates

The radiation doses at the site boundary and off the site that could have been received by the public from radioactive materials and radiation leaving the site were calculated. These calculations are made for four exposure pathways; submersion, inhalation, ingestion, and direct radiation from external sources. DOE interim guidance⁶ requires the use of the EPA-AIRDOSE/RADRISK model and computer program¹⁶ for the calculation of the submersion dose for radionuclides released to the air. The inhalation and ingestion pathway calculations require the use of the DOE interim 50-year Committed Dose Equivalent Factors.⁶

a. Submersion Pathway

The EPA-AIRDOSE/RADRISK computer code uses a modified Gaussian plume equation to estimate both horizontal and vertical dispersion of radionuclides released to the air from stacks or area sources. For 1986, submersion dose calculations were carried out for carbon-11, argon-41, krypton-85, and radon-220. The annual release rates are those listed in Table 7. The wind speed and direction data shown in Figure 3 are the meteorological data needed for these calculations. The calculations were carried out to 80 km (50 mi) using the population distribution of 16 segments and ten distant increments given in Table 1. The dose rate was calculated at the midpoint of each interval and integrated over the entire area to give the annual cumulative population dose.

The highest perimeter dose rates are in the north to east sectors. The closest full-time resident, who would receive the largest dose, is located approximately 0.5 km (0.3 mi) north of the site boundary. The results are summarized in Table 22. The applicable DOE dose limits to members of the public are 500 mrem/y for occasional annual exposure (less than five years) and 100 mrem/y (more than five years) for continuous exposure.

TABLE 22

Submersion Dose From Airborne Emissions, 1986
(Millirems)

Nuclide	Quantity Released (Ci)	Maximum Perimeter	Maximum Individual
		50-Year	50-Year
Carbon-11	90	0.038	0.011
Argon-41	1.5	0.0010	0.0003
Krypton-85	1.7	0.0073	0.0026
Radon-220	6982	0.352	0.017

The population data in Table 1 was used to calculate the cumulative population dose from gaseous radioactive effluents. The results are given in Table 23, together with the natural external radiation dose. The natural radiation dose was that measured at the off-site TLD locations, and it is assumed that this dose is representative of the entire area within an 80 km (50 mi) radius.

TABLE 23

80 km Population Dose, 1986	
Source	Man-Rems
	50-Year
Carbon-11	0.36
Argon-41	0.03
Krypton-85	0.37
Radon-220	0.10
Natural	6.20×10^5

b. Inhalation Pathway

The potential radiation exposures by the inhalation pathways were calculated by the methodology specified in the interim guidance.⁶ The total quantity for each radionuclide inhaled, in μCi , is calculated by multiplying the annual average air concentrations by the general public breathing rate of $8400 \text{ m}^3/\text{y}$.¹⁷ This annual intake is then multiplied by the EFF.D.E. for the appropriate lung retention class (D, W, or Y). Because the EFF.D.E. are in units of $\text{Rem}/\mu\text{Ci}$, this calculation gives the 50-year committed effective dose equivalent directly. The applicable EFF.D.E.s are collected in Table 24.

The calculated doses in Tables 4, 5, 6, and 8 were obtained using this procedure. Because they are all essentially perimeter locations, these doses represent the fenceline values for those radionuclides measured.

In almost all cases, these doses also are the same as the off-site measurements and represent the ambient dose for the area from these nuclides. Doses are not calculated for the total alpha and total beta measurements since the interim guidance does not provide EFF.D.E.s for such measurements.

The other source of inhalation exposure is from the tritiated water vapor discharged from the stacks. The annual release rates and locations are given in Table 7. The concentrations at various distances and doses were calculated by a computer program based on an atmospheric dispersion model^{18,19} and the hydrogen-3 EFF.D.E. from Table 24. The doses were calculated for the same population group as for the submersion pathway and are collected in Table 25.

c. Ingestion Pathway

Similarly, following the methodology outlined in the interim guidance, the ingestion annual intake, in μCi , is obtained by multiplying the concentration ($\mu\text{Ci/mL}$) by the annual water consumption of a member of the general public (7.3×10^5 mL). This annual intake is then multiplied by the EFF.D.E. for ingestion (Table 24) to obtain the dose. This is carried out for all radionuclides and summed to obtain the total ingestion dose.

The only location where radionuclides attributable to Argonne operations could be found in off-site water was Sawmill Creek below the wastewater outfall. Although this water is not used for drinking purposes, the 50-year effective dose equivalent was calculated for a hypothetical individual ingesting water at the concentrations found. Those nuclides added to Sawmill Creek by Argonne waste water, their net concentrations in the Creek (16K results subtracted) and the corresponding dose rates, if water at these concentrations were used as the sole water supply by an individual, are given in Table 26. The dose rates were all well below the standards for the general population. It should be emphasized that Sawmill Creek is not used for drinking, swimming, or boating. Inspection of the area shows there are few fish in the stream, and they do not constitute a significant source of food for any individual.

TABLE 24

50-Year Committed Dose Equivalent Factors - EFF.D.E.
(Rem/ μ Ci)

Nuclide	Ingestion	Inhalation
Hydrogen-3	6.3×10^{-5}	6.3×10^{-5}
Beryllium-7	-	2.7×10^{-4}
Carbon-11	-	8.0×10^{-6}
Strontium-89	-	0.037
Strontium-90	0.13	1.3
Lead-210	-	0.13
Radium-226	1.1	-
Thorium-228	-	0.031
Thorium-230	-	0.062
Thorium-232	-	0.0011
Uranium-234	0.26	0.013
Uranium-235	0.25	0.012
Uranium-238	0.23	0.012
Neptunium-237	0.39	-
Plutonium-238	0.38	-
Plutonium-239	0.43	0.033
Americium-241	2.2	-
Curium-242	0.065	-
Curium-244	1.1	-
Californium-249	2.3	-
Californium-252	0.48	-

TABLE 25

Hydrogen-3 Inhalation Dose, 1986

Maximum Perimeter	0.040 mrem
Maximum Individual	0.014 mrem
80 km Population Dose	2.03 man-rem

TABLE 26

Radionuclide Concentrations and Dose Estimates
for Sawmill Creek Water, 1986

Nuclide	Total Released (Millicuries)	Net Avg. Conc. (10^{-9} μ Ci/mL)	Dose (mrem/year)
Hydrogen-3	3.12×10^4	2525	0.116
Strontium-90	43	3.46	0.328
Neptunium-237	0.01	0.0010	0.0003
Plutonium-239	0.02	0.0018	0.0006
Americium-241	0.14	0.0112	0.0180
Sum			0.463

As indicated in Table 9, occasional Sawmill Creek samples (less than 10) contained traces of plutonium-238, curium-242,244, or californium-249,252, but the averages were only slightly greater than the detection limit. The annual dose due to an individual consuming water at these concentrations can be calculated as was done for those radionuclides more commonly found in Creek water, but the method of averaging probably overestimates the true concentration. These annual doses range from 3×10^{-3} to 6×10^{-5} mrem/y for these radionuclides.

The U. S. Environmental Protection Agency (EPA) has established drinking water standards based on a dose of 4 mrem/y for man-made beta particle and photon emitting radionuclides.¹² The EPA standard is 2×10^{-5} μ Ci/mL for hydrogen-3 and 8×10^{-9} μ Ci/mL for strontium-90. The concentrations in Table 26 correspond to 13% (hydrogen-3) and 43% (strontium-90) of the EPA standards. No specific EPA standards exist for the transuranic nuclides.

d. External Direct Radiation Pathway

The TLD results in Section III.A.6. are used to calculate the radiation dose from external sources. Above-normal fence-line doses attributable

to Argonne operations were found at the south boundary near the Waste Storage Facility (Location 7I) and at the north boundary near Building 202 (Location 14I).

At Location 7I, the above-background fenceline dose from Argonne was about 366 mrem/y. Approximately 300 m (0.3 mi) south of the fenceline (grid 6I), the measured dose was 78 ± 9 mrem/y, well within the normal range of the off-site average (78 ± 5 mrem/y). There are no individuals living in this area. The closest residents are about 1.6 km (1 mi) south of the fenceline. At this distance, the calculated dose rate from the Waste Storage Area is about 0.02 mrem/y, if the energy of the radiation were the cesium-137 0.66 MeV gamma-ray and about 0.08 mrem/y if the energy were the 1.33 MeV cobalt-60 gamma-ray. In the area north of the site, the fenceline radiation dose from the cobalt-60 sources in Building 202 was measured at 24 mrem/y. The nearest residents are 750 m (0.47 mi) to the north-northwest. The calculated dose at that location was about 0.11 mrem/y.

At the fenceline, where higher doses were measured, the land is wooded and unoccupied. All of these dose calculations are based on full-time, outdoor exposure. Actual exposures to individuals are substantially less since they are inside (which provides shielding) or away from their dwelling some of the time.

e. Dose Summary

The total dose received by off-site residents was a combination of the separate pathways that contribute to this total: carbon-11, argon-41, krypton-85, and radon-220 submersion dose; hydrogen-3 inhalation dose; and cobalt-60 external radiation dose. The highest dose was about 0.15 mrem/y to individuals living north of the site if they were outdoors at that location during the entire year. The total annual population dose to the entire area within an 80 km (50 mi) radius is 2.9 man-rem.

B. Chemical Constituents

1. Surface Water

The environmental water data contained in this section were collected to ascertain the effectiveness of Argonne's effluent pollution controls and determine compliance with State regulations and National Pollutant Discharge Elimination System (NPDES) permits. The appropriate stream and effluent standards are listed in the STATE OF ILLINOIS RULES AND REGULATIONS, Title 35, Subtitle C, Chapter I.²⁰ Stream Quality Standards appear in Part 302, Subpart B and Effluent Quality Standards appear in Part 304, Subpart A of that document. All of the results are compared to the State standards, which are listed in Table 27. Minimum detectable amounts (detection limits) are included for comparison. The detection limits for atomic absorption methods represent twice the background variation, which is commonly used for this purpose. Detection limits for ion selective methods are those listed by the manufacturer. The lower limit of detection for x-ray fluorescence is based on the appropriate counting statistics.

The analytical methods have been described in previous reports in this series. The results of measurements for chemical constituents are expressed as milligrams (mg) or micrograms (μg) per liter (L). Averages were calculated as described in Section III.A. Yearly averages are reported with a (+) limit value. This value is the standard error at the 95% confidence limit and is calculated from the standard deviation of the yearly average. Only when sample concentrations are random does this value represent the actual distribution occurring at the sampling location. As in the past, emphasis has been placed on Sawmill Creek, since this is the principal route for waste water leaving the Argonne site, and on the NPDES permit locations.

a. Waste Water Treatment Plant Effluent

The major discharge of waste water from Argonne operations is by way of the waste water treatment plant. The water volume from this source was approximately 2.8 megaliters (0.74 million gallons) per day in 1986 and was

TABLE 27

Water Quality Standards and Detection Limits
(Concentrations in mg/L)

Constituent	State Standard		Detection Limit
	Stream	Effluent	
Ammonia Nitrogen (as N)	1.5	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	0.1
Arsenic	1.0	0.25	0.005
Barium	5.0	2.0	0.002
Cadmium	0.05	0.15	0.0002
Chloride	500	-	1.0
Chromium	1.0	1.0	0.001
Copper	0.02	1.0	0.001
Cyanide	0.025	0.025	0.020
Fluoride	1.4	15	0.02
Iron	1.0	2.0	0.10
Lead	0.1	0.1	0.001
Manganese	1.0	1.0	0.001
Mercury	0.0005	0.0005	0.00005
Nickel	1.0	1.0	0.003
pH	6.5-9.0	6.0-9.0	-
Selenium	1.0	-	0.005
Silver	0.005	0.1	0.0002
Sulfate	500	-	1.0
Total Dissolved Solids	1000	-	-
Zinc	1.0	1.0	0.01

comprised of 48% sanitary waste water and 52% water from laboratory operations. Laboratory waste water is directed to a holding pond with an outflow of about 1300 liters/minute (350 gallons/minute). The discharge occurs over a 24-hour period. Since the sanitary system is fairly constant, the overall output is relatively constant.

Water from the waste water treatment plant is sampled on a continuous basis during the work week. A flow proportional 24-hour sample of the combined sanitary and laboratory effluent is obtained each day and analyzed for constituents of interest.

Past experience has shown that levels of arsenic, barium, beryllium, cadmium, lead, nickel, selenium, silver, and zinc in the effluent were normally 5% of the State limits. In addition, the analytical sensitivity is such that very low levels of these constituents can be determined. On this basis, effluent samples to be analyzed for these constituents are composited on a monthly basis. If results approached the State limit, the individual samples could be analyzed to determine if the value of any sample was equal to the limit.

The results obtained for chemical constituents in the effluent water from the treatment plant are shown in Table 28. All of the average concentrations are below the State standards. The average value for mercury is 29% of the State standard and 4% of the samples exceeded this value. The corresponding values for 1985 were 60% and 12%. Efforts are continuing to lower mercury levels by monitoring the waste water retention tanks most likely to contain mercury, and treating those tanks containing elevated concentrations. This year's results indicate a substantial improvement over previous years. Concentrations of other constituents are similar to last year's values.

b. National Pollutant Discharge Elimination System (NPDES)

The Laboratory discharges are regulated by NPDES Permit #IL 0034592. This permit is subject to renewal about every three to five years. Effective October 1984, this permit was renewed to March 1, 1989, and several

TABLE 28
 CHEMICAL CONSTITUENTS IN EFFLUENTS FROM ANL TREATMENT PLANT, 1986
 (CONCENTRATIONS IN MICROGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.	PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
ARSENIC	12	-	-	< 5	< 2.0	0
BARIUM	12	17 ± 6	7	39	0.8	0
BERYLLIUM	12	0.03 ± 0.01	0.01	0.08	-	-
CADMIUM	12	0.5 ± 0.3	< 0.2	1.6	0.3	0
CHROMIUM	52	7.8 ± 1.4	2	35	-	-
COPPER	52	22 ± 3	10	65	2.2	0
FLUORIDE	12	330 ± 21	272	388	2.2	0
IRON	52	187 ± 42	50	1210	9.4	0
LEAD	12	2.5 ± 0.6	< 2.0	4.1	2.5	0
MANGANESE	52	51 ± 16	10	374	5.1	0
MERCURY	52	0.15 ± 0.04	0.05	0.97	30	4
NICKEL	12	29 ± 13	2	57	2.9	0
PH	249	-	6.9	8.3	-	0
SELENIUM	12	-	-	< 5	< 0.5	0
SILVER	12	2.6 ± 0.4	1.4	3.6	2.6	0
ZINC	12	59 ± 14	33	103	5.9	0

revisions have been made. The locations previously described¹³ and shown in Figure 6, remain the same. For purposes of clarity, the location numbers in the figure are given without the zeros. Thus, permit Location 001 is shown in the figure as 1. The location formerly denoted as 001 (waste water treatment plant) has been divided into 001A (sanitary waste) and 001B (laboratory waste). Requirements for the combined effluent are still listed under Location 001.

The permit was modified in January 1986 to provide for bimonthly collection of samples in Sawmill Creek upstream and downstream of the Argonne outfall. The samples are analyzed for total dissolved solids and chloride content. The purpose of these studies is to establish a background for these constituents prior to the change in Argonne's domestic water treatment procedure in November 1986. A coagulation procedure was partially replaced by ion exchange in November and the changeover was completed in December. Results are listed in Table 29 so that a comparison may be made to previous years. The results for the first eleven months of the year are similar to previous years. The total dissolved solids and chloride levels upstream exceeded the downstream levels by about 20%. In the last half of November, there was a dramatic change in the distribution of these materials. This change resulted from a combination of the cessation of operation at the Marion Brook Waste Treatment Plant, which had released high levels of both chloride and total dissolved solids, and the beginning of the ion exchange process which increased Argonne's effluent levels of these constituents substantially. The chloride level was higher downstream by about a factor of three and the total dissolved solids level was about a factor of two higher.

Samples at Locations 001A, 001B, and 001 are collected weekly, while all other samples are collected monthly. Results for 1986 are in Table 29. Total suspended solids occasionally exceeded the permit levels at several locations, usually when excessive rainfall occurred. One sample exceeded the limit for fecal coliform, presumably due to low residual chlorine levels. The water discharged at Location 009 always had a pH greater than the limit of 9.0 and frequently had excessive suspended solid levels. The effluent point is comprised of overflow water from a holding pond

TABLE 29

NPDES Effluent Quality Summary, 1986

Discharge Location	Permit Constituent	Concentration Limits mg/L		Number Exceeding Limit	* _R <u>Measured Permit</u>
		30 Day Average	Daily Max.		
001A	Flow	None		0	-
	BOD	30	60	0	-
	TSS	30	60	1	1.5
001B	Flow	None		0	-
	Chemical Oxygen Demand	-	-	0	-
	TSS	15	30	1	1.3
	Mercury	0.003	0.006	0	-
001	pH	6-9		0	-
	Fecal Coliform	- <u>400 organisms</u> 100 mL		1	3.2
	BOD	30	60	0	-
	TSS	30	60	0	-
	Temperature	< 2.8°C Rise		0	-
002	Flow	None		0	-
	pH	6-9		0	-
	TSS	15	30	0	-
	Temperature	< 2.8°C Rise		0	-
003	Flow	None		0	-
	pH	6-9		0	-
	TSS	15	30	3	1.2-3.4
	Temperature	< 2.8°C Rise		0	-

TABLE 29 (contd)

Discharge Location	Permit Constituent	Concentration Limits mg/L		Number Exceeding Limit	* _R <u>Measured</u> / <u>Permit</u>
		30 Day Average	Daily Max.		
004	Flow	None		0	-
	pH	6-9		0	-
	TSS	15	30	3	1.1-3.3
	Temperature	< 2.8°C Rise		0	-
005	Flow	None		0	-
	pH	6-9		0	-
	Temperature	< 2.8°C Rise		0	-
	Fats, Oil, & Grease	15	30	0	-
006	Flow	None		0	-
	pH	6-9		0	-
	TSS	15	30	2	1.4-8.2
	Zinc	1.0	2.0	0	-
007	Flow	None		0	-
	pH	6-9		2	9.1, 9.1
	Temperature	< 2.8°C Rise		0	-
008	Flow	None		0	-
	pH	6-9		0	-
009	Flow	None		0	-
	pH	6-9		11	9.7-11.5
	TSS	15	30	5	1.4-6.2

* R is the range of the ratio of the values of the measurements exceeding the concentration limit to the concentration limit (except for pH, for which the actual values are given).

associated with the coagulation procedure which has been discontinued resulting in no further discharge. Water levels in the pond are checked to assure that precipitation does not cause overflow.

c. Sawmill Creek

Samples collected for evaluation of the effect of the sanitary waste on stream quality are obtained semimonthly using specially constructed sampling bottles. The sampling bottles are designed to provide temperature measurements as well as to minimize changes in oxygen content during collection. These samples are collected 15 m (50 ft) upstream of the Argonne wastewater outfall [7M (up)] and 100 m (310 ft) downstream of the outfall [7M (down)]. The data from these studies are in Table 30. Average levels for ammonia nitrogen are below the State limit at both locations. The dissolved oxygen levels obtained during 1986 are all above the State minimum of 5 mg/L. Three individual samples exceeded the State limit for cyanide, but the source is unknown.

Samples to evaluate the effect of combined sanitary and laboratory waste on the concentrations of chemical constituents in Sawmill Creek are collected by a proportional sampler. These are the same samples taken for radioactivity analyses. The results are summarized in Table 31.

None of the individual samples exceeded the State stream standards for arsenic, barium, cadmium, chromium, fluoride, lead, manganese, nickel, selenium, silver, and zinc. As in the past, high iron levels reflect high turbidity conditions and are not related to Argonne releases. The level of copper exceeded State stream quality limits in 23% of the samples, but this is not due to Argonne operations because the average stream and effluent copper concentrations differ by a factor of two, but the effluent water is diluted by about a factor of ten with stream water. The copper is believed to be of natural origin. One sample exceeded the mercury State stream standard of 0.5 µg/L by 20%.

TABLE 30

SAWMILL CREEK - EFFECT OF SANITARY WASTE, 1986

CONSTITUENT	* LOCATION	NO. OF SAMPLES	CONCENTRATION (MG/L)			AVG. PERCENT OF STANDARD	PERCENT EXCEEDING STATE STANDARD
			AVG.	MIN.	MAX.		
AMMONIA NITROGEN	7M (UP)	24	0.3 ± 0.1	0.1	1.3	19	0
	7M (DOWN)	24	0.5 ± 0.3	0.1	3.5	32	4
CHLORIDE	7M (UP)	24	316 ± 66	116	616	-	-
	7M (DOWN)	24	274 ± 51	91	520	-	-
CYANIDE	7M (UP)	3	-	-	< 0.02	< 80	0
	7M (DOWN)	24	0.02 ± 0.01	< 0.02	0.07	80	13
DISSOLVED OXYGEN	7M (UP)	24	11.3 ± 0.8	7.7	15.3	-	0
	7M (DOWN)	24	10.8 ± 0.6	7.7	13.3	-	0
DISSOLVED SOLIDS	7M (UP)	24	1040 ± 160	460	1600	-	-
	7M (DOWN)	24	940 ± 124	445	1340	-	-
** PH	7M (UP)	24	-	7.7	8.8	-	0
	7M (DOWN)	24	-	7.7	8.8	-	0
SULFATE	7M (UP)	24	151 ± 22	68	237	30	0
	7M (DOWN)	24	166 ± 18	72	241	33	0
*** TEMPERATURE	7M (UP)	24	14.0 ± 3.4	0.4	26.0	-	-
	7M (DOWN)	24	15.2 ± 2.9	2.4	25.7	-	-

* LOCATION 7M (UP) IS 15 M (50 FT) UPSTREAM FROM THE WASTE-WATER OUTFALL. ALL OTHER SAMPLES WERE COLLECTED 60 M (200 FT) DOWNSTREAM FROM THE OUTFALL.

**
UNIT

DEGREES CENTIGRADE

TABLE 31
 CHEMICAL CONSTITUENTS IN SAWMILL CREEK LOCATION 7M, 1986*
 (CONCENTRATIONS IN MICROGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.	PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
ARSENIC	12	-	-	< 5	< 0.5	0
BARIUM	12	53 ± 15	21	97	1.1	0
BERYLLIUM	12	0.05 ± 0.03	0.01	0.18	-	-
CADMIUM	12	0.7 ± 0.2	0.2	1.6	1.5	0
CHROMIUM	52	8 ± 2	3	37	-	-
COPPER	52	17 ± 2	9	45	85	23
FLUORIDE	12	487 ± 73	280	670	35	0
IRON	52	698 ± 167	193	3670	70	15
LEAD	12	4 ± 1	1	8	3.9	0
MANGANESE	52	81 ± 12	18	285	8.1	0
MERCURY	52	0.09 ± 0.03	< 0.05	0.58	19	2
NICKEL	12	32 ± 14	2	62	3.2	0
PH	248	-	7.1	8.9	-	0
SELENIUM	12	-	-	< 5	< 0.5	0
SILVER	52	1.7 ± 0.2	0.2	3.1	33	0
ZINC	12	32 ± 7	10	45	3.2	0

* LOCATION 7M IS 60 M (200 FT) DOWNSTREAM FROM THE WASTE-WATER OUTFALL.

d. Des Plaines River

The effect of Sawmill Creek on the Des Plaines River is evaluated by collecting samples at Willow Springs (upstream of Argonne) and at Lemont (downstream of Argonne). These samples are analyzed for total mercury and all results are less than the detection limit of 0.05 $\mu\text{g/L}$. In no case is there any indication that mercury levels are affected by the Argonne effluent.

2. Ground Water

A sanitary landfill for non-radioactive waste is located on the western edge of the Argonne site (Figures 1 and 7). This landfill operates under Illinois EPA permit No. 1981-29-OP and is further described in Reference 13. Operation of the ANL landfill began in July, 1966. During the period 1969-1978, substantial quantities of liquid organic and inorganic wastes were disposed of on-site by adding them to an open "French drain" located in the northeast sector of the landfill.²

In 1979, an investigation was begun to determine the subsoil characteristics of the site and to place ground-water monitoring wells at appropriate points in and around the landfill. Because the topography suggested a southerly water flow, Wells 1 and 5 were located outside the landfill and were meant to measure water entering and leaving the landfill. Wells 2, 3, and 4 were placed in the landfill itself. In April, 1980,²¹ a more comprehensive study was initiated. Three additional test wells were placed in the landfill at previously untested locations. Well 6 was placed in the east section to sample any water flowing out of the landfill in the southeasterly direction. Wells 7a and 7b were located along the south side and were nested. They were installed to measure vertical water movement as well as to provide monitoring of water from two depths.

The study established the following important characteristics. There is a perched water condition at a depth varying from about 6 m (20 ft) on the north to about 7.6 m (25 ft) on the south. This is caused by the relatively impermeable condition of the underlying clay which restricts downward water flow. There are several granular seams in the soil above the

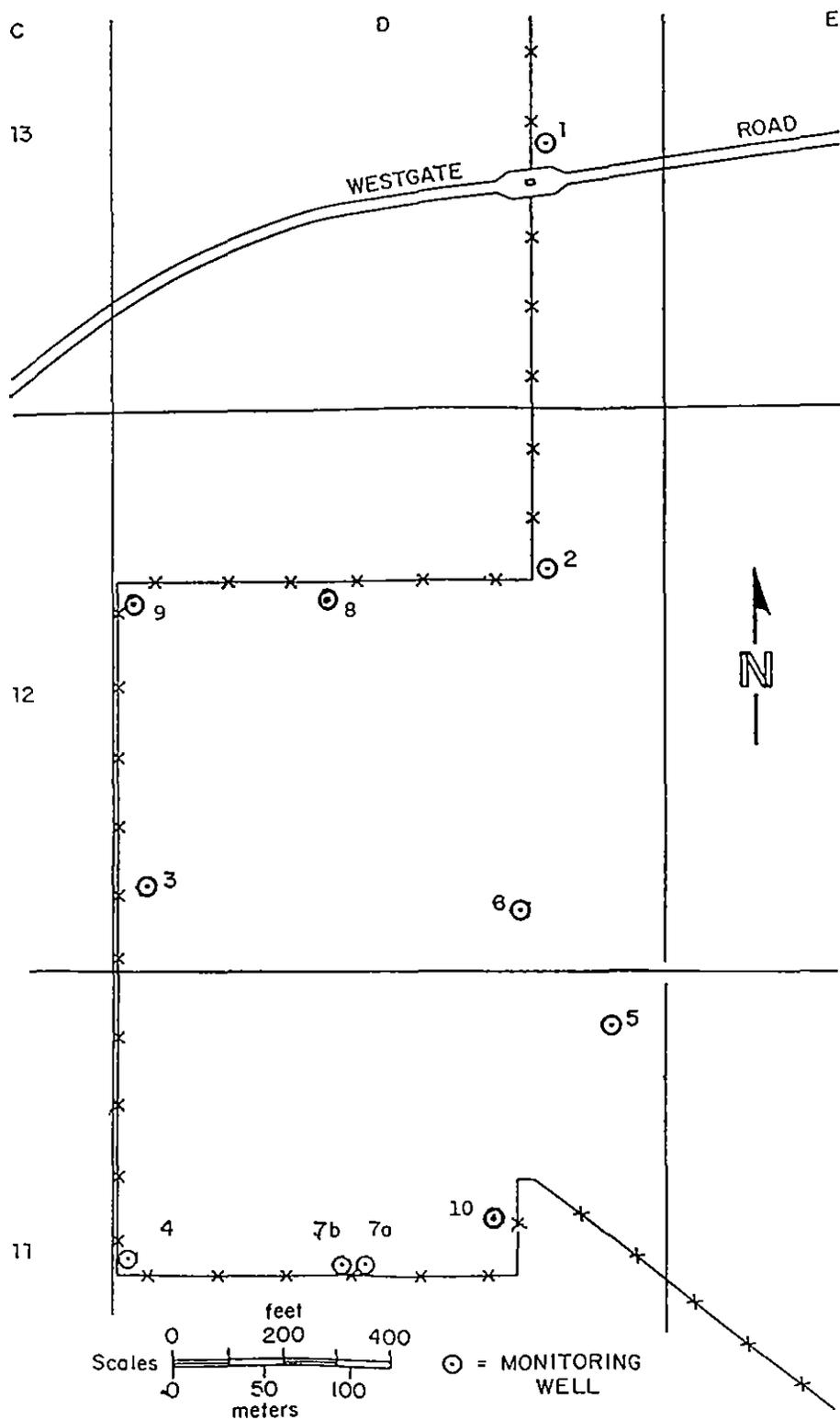


Fig. 7. Monitoring Well Locations for Landfill

clay which permit lateral flow. Additionally, regional ground-water flow patterns confirm that the flow is generally southerly.

These facts indicate that water in the test wells is from surface infiltration and that it moves horizontally approximately 7.6 m (25 ft) below the surface. Penetration to the dolomite aquifer used for water supplies, 15-30 m (50-100 ft) below the surface, is very slow. Indeed, core samples obtained at 11-12 m (35-40 ft) below the surface were only moist indicating that the aquifer is below this level and little downward movement is occurring. If movement was occurring, this stratum would be nearly saturated. In September, six new wells were constructed. Wells 1, 2, and 4 were suspected of being poorly sealed and replacement wells were drilled (1-2, 2-2, and 4-2) within five feet of the originals. Additional wells, 8, 9, and 10 (see Figure 7) were constructed to improve the peripheral coverage.

The original wells have been studied from 1979 until the present to determine constancy of water levels, recharge rates, and chemical characteristics. The new wells have been sampled in October and December of this year. The water levels, measured quarterly, for 1986 are shown in Table 32. In addition, the precipitation for a 30-day period prior to sampling and the average levels for 1981-1985 are shown. The average water levels for Wells 1-4 were similar in 1986 to the 1981-1985 average. The average water levels in Wells 6 and 7a were higher in 1986 than the 1981-1985 average. However, little change in water levels of Wells 6 and 7a occurred between 1985 and 1986.

The samples for analyses were obtained after removing and discarding at least one well volume and allowing the water level to re-establish. The recharge rates are variable and may take up to one week, although some wells take less than 24 hours. Water removal is accomplished using a special small diameter submersible pump.

On the basis of past data, selected elements were studied in the original wells, while the new wells were analyzed for the full spectrum of elements. For the following discussion the replacement wells will be compared

TABLE 32

Well Point and Water Level Elevations⁽¹⁾ of
Monitoring Sites at the ANL Landfill - 1986

Well No.	Ground Surface Elevation (3)	Well Point Elevation	Quarterly Measurements				Average	
			1st	2nd	3rd	4th	1986	1981-85
1	227.53	218.53	227.11	226.16	-	-	226.64	226.45
1-2	227.53	219.91	-	-	221.38	221.96	221.61	-
2	230.58	220.83	225.64	225.25	-	-	225.45	225.58
2-2	230.58	214.73	-	-	225.06	225.98	225.52	-
3	226.77	218.08	224.30	224.18	223.85	224.12	224.11	224.33
4	227.23	221.13	225.37	225.16	-	-	225.27	225.08
4-2	227.23	220.22	-	-	222.20	224.49	223.35	-
5	227.53	215.34	*	*	217.17	221.71	219.44	220.67
6	229.91	215.13	220.89	225.19	224.73	224.73	223.89	221.51
7a	227.81	220.19	226.50	223.97	225.61	226.31	225.60	223.46
7b	227.81	214.09	**	**	**	**	**	**
8	230.73	222.20	-	-	225.70	226.22	225.96	-
9	229.51	223.72	-	-	226.28	226.68	226.48	-
10	228.90	222.50	-	-	227.35	228.42	227.89	-
(2)	-	-	48.8	84.1	193.3	58.7	96.2	67.0

* Not measured.
** Dry.

- Notes: (1) All measurements are in meters above mean sea level.
(2) Precipitation (water equivalent) in millimeters for 30-day period prior to measurements. Data for 3rd and 4th quarters were obtained from NOAA as measured at O'Hare International Airport. Data from ANL not available after June.
(3) The ground surface elevations of Wells 1-7b are as previously reported, while those of Wells 8, 9, and 10 are estimated, pending completion of a resurvey of the entire landfill area.

to the original wells. Well 1 (Table 33) has had persistently high levels of chloride and dissolved solids in the past and in 1986. The levels of chloride and dissolved solids in Well 1-2 (Table 34) were lower. The manganese in Well 1-2 was higher by about a factor of two. The pH in Well 1-2 was considerably higher and thus the iron is considerably lower.

Well 2 (Table 35) has had a pH of 11 or greater from the beginning of sampling until the last two years. This was presumably due to the nearby disposal of carbonate-type water softening materials. The high pH water was confined to this area since no other wells have had elevated or changing pH values. The pH of samples obtained in 1986 were 7.8 and 7.9. This indicates the continued depletion of the source of basicity. Well 2-2 (Table 36) was drilled in close proximity to Well 2. The pH of the samples obtained were both 12, thus indicating that Wells 2 and 2-2 are in an area where carbonate-type materials were disposed. It is interesting to note that even though these wells are close to one another, there is apparently no interaction between them in the water layer since the hydrogen ion concentration differs by a factor of about 10^4 . The other differences can be seen by comparing the respective tables.

Well 3 (Table 37) is characterized by the presence of arsenic but at much lower levels than in previous years. The levels have changed regularly from a high of 35 $\mu\text{g/L}$ in 1983 to current levels of about 7 $\mu\text{g/L}$, which is near the detection limit. The source of the arsenic is unknown and the results indicate depletion is occurring by dilution. Arsenic has not been found in any other well, until traces were detected this year in Well 9.

Well 4 (Table 38) continues to show high levels of manganese and sulfate but relatively low iron levels. This well has a fast recharge rate and is located close to a ditch which drains a marshy area. This ditch was sampled in 1984, once in 1985, and once in 1986. No unusual values were seen with the exception of an elevated chloride level which could be related to runoff from the road. Both manganese and sulfate levels are low indicating little or no connection between the ditch and Well 4. Well 4-2 (Table 39) has similar chemical characteristics to Well 4 with the notable

TABLE 33
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 1
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	163 ± 490	49	277
CHLORIDE *	2	1060 ± 200	1020	1110
DISSOLVED SOLIDS *	2	2570 ± 670	2410	2720
FLUORIDE	2	155 ± 30	148	162
IRON	2	749 ± 389	659	840
MANGANESE	2	170 ± 58	157	184
MERCURY	2	-	-	< 0.05
PH	2	-	7.0	7.1
SELENIUM	2	-	-	< 5
SULFATE *	2	145 ± 101	121	168
TEMPERATURE **	2	11.2	9.8	12.7

*
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

TABLE 34
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 1-2
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	180 ± 351	99	262
CADMIUM	2	3.10 ± 5.59	1.80	4.40
CHLORIDE *	2	659 ± 37	650	667
COPPER	2	21 ± 5	20	22
DISSOLVED SOLIDS *	2	1870 ± 410	1780	1970
FLUORIDE	1	-	-	172
IRON	2	-	-	< 100
LEAD	2	2 ± 5	1	3
MANGANESE	2	337 ± 373	250	424
MERCURY	2	-	-	< 0.05
NICKEL	2	138 ± 101	114	161
PH	2	-	7.6	7.8
SELENIUM	2	-	-	< 5
SILVER	2	1.50 ± 4.73	0.40	2.60
SULFATE *	2	137 ± 47	126	148
TEMPERATURE **	2	12.3	11.4	13.2
ZINC	2	20	20	20

* CONCENTRATION IN MILLIGRAM/LITER.

** DEGREES CENTIGRADE

TABLE 35
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 2
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	254 ± 63	240	269
CHLORIDE *	2	27 ± 2	26	27
DISSOLVED SOLIDS *	2	363 ± 13	360	366
FLUORIDE	2	150 ± 17	146	154
IRON	2	-	-	< 100
MANGANESE	2	31 ± 12	28	34
MERCURY	2	-	-	< 0.05
PH	2	-	7.8	7.9
SELENIUM	2	-	-	< 5
SULFATE *	2	75	75	75
TEMPERATURE **	2	11.9	11.6	12.3

*
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

TABLE 36
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 2-2
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	569 ± 253	510	628
CADMIUM	2	0.25 ± 0.22	< 0.20	0.30
CHLORIDE *	2	58 ± 103	34	82
COPPER	2	7 ± 15	3	10
DISSOLVED SOLIDS *	2	691 ± 209	642	739
FLUORIDE	1	-	-	282
IRON	2	148 ± 207	100	196
LEAD	2	2 ± 3	1	2
MANGANESE	2	3 ± 1	3	3
MERCURY	2	-	-	< 0.05
NICKEL	2	21 ± 11	18	23
PH	2	-	12	12
SELENIUM	2	-	-	< 5
SILVER	2			
SULFATE *	2	28 ± 13	25	31
TEMPERATURE **	2	12.1	11.0	13.2
ZINC	2	20	20	20

* CONCENTRATION IN MILLIGRAM/LITER.

** DEGREES CENTIGRADE

TABLE 37
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 3
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	4	7 ± 5	< 5	13.0
BARIUM	4	220 ± 187	94	367
CHLORIDE *	4	6 ± 7	1	12
DISSOLVED SOLIDS *	4	796 ± 53	766	852
FLUORIDE	3	141 ± 15	136	150
IRON	4	1090 ± 2490	50	3780
MANGANESE	4	203 ± 29	174	224
MERCURY	4	-	-	< 0.05
PH	4	-	6.7	6.8
SELENIUM	4	-	-	< 5
SULFATE *	4	81 ± 65	42	139
TEMPERATURE **	4	11.6	10.8	12.5

* CONCENTRATION IN MILLIGRAM/LITER.

** DEGREES CENTIGRADE

TABLE 38
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 4
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	122 ± 248	64	179
CADMIUM	1	-	-	0.80
CHLORIDE *	2	271 ± 364	186	355
COPPER	1	-	-	6
DISSOLVED SOLIDS *	2	1200 ± 500	1090	1320
FLUORIDE	2	206 ± 26	200	212
IRON	2	508 ± 127	479	538
LEAD	1	-	-	4
MANGANESE	2	1350 ± 1380	1030	1670
MERCURY	2	-	-	< 0.05
NICKEL	1	-	-	35
PH	2	-	6.8	6.9
SELENIUM	2	-	-	< 5
SILVER	1	-	-	0.50
SULFATE *	2	212 ± 269	149	274
TEMPERATURE **	2	10.3	8.0	12.7
ZINC	1	-	-	20

*
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

TABLE 39
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 4-2
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	47 ± 66	32	62
CADMIUM	2	0.45 ± 0.65	0.30	0.60
CHLORIDE *	2	45 ± 34	37	53
COPPER	2	13 ± 22	8	18
DISSOLVED * SOLIDS	2	881 ± 454	775	986
FLUORIDE	1	-	-	304
IRON	2	-	-	< 100
LEAD	2	1 ± 1	1	1
MANGANESE	2	10 ± 15	6	13
MERCURY	2	-	-	< 0.05
NICKEL	2	40 ± 51	28	52
PH	2	-	8.0	9.0
SELENIUM	2	-	-	< 5
SILVER	2	1.00 ± 0.43	0.90	1.10
SULFATE *	2	146 ± 127	116	175
TEMPERATURE **	2	11.0	10.2	11.8
ZINC	2	20	20	20

*
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

exception of manganese, iron, and pH. The much higher pH value of 4-2 (average 8.5) as compared to the pH value of Well 4 (average 6.9) probably accounts for this difference. Once again two wells in close proximity have differences in characteristics.

Well 6 (Table 40) contains high levels of iron and manganese and moderate to high levels of chloride and sulfate. The elevated concentrations are possibly due to manganese sulfate and ferric chloride disposed of in the landfill during previous years. Results from Well 7a (Table 41) and Well 5 (Table 42) are about the same as in past years with no outstanding characteristics.

New Wells 8 (Table 43) and 9 (Table 44) are located along in the northern boundary of the landfill. The most noteworthy characteristic is the presence of low levels of arsenic in Well 9. This well is located in the general area of Well 3 which has shown persistent, but higher, levels in the past. This would indicate some linkage between these wells or the presence of arsenic in a location adjacent to them. However, the manganese and chloride levels are higher in Well 9 than in Well 3 by at least a factor of ten. Chloride is generally considered to be mobile and this large difference would not exist if they were interconnected. A trace of mercury has been found in one of two samples obtained from Well 9. Well 10 (Table 45) is located between Wells 6 and 7a and more closely resembles Well 7a.

The most characteristic factor in the results is the constancy of the pH values at each well. With the exception of Well 2, the pH values of each well tested in 1986 are the same as 1985 values within 0.1 pH unit. The magnitude of the values were different from each other, except for Well 3 and Well 9 which were the same. It appears that some buffering system is available in each which maintains the pH. The new wells have not been sampled sufficiently, but the presence of arsenic in Well 9 and the high pH in Well 2-2 is of interest. In addition, arsenic is also found in Well 3 at about the same level. Manganese in Wells 4, 6, and 9 is high and fairly constant. It does not appear that water is moving from one well site to another and some type of equilibrium exists in each well.

TABLE 40
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 6
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	4	-	-	< 5
BARIUM	4	138 ± 116	61	256
CHLORIDE *	4	251 ± 45	215	294
DISSOLVED SOLIDS *	4	1310 ± 39	1280	1350
FLUORIDE	3	137 ± 38	114	154
IRON	4	8640 ± 9170	50	15100
MANGANESE	4	2910 ± 450	2490	3230
MERCURY	4	-	-	< 0.05
PH	4	-	6.4	6.6
SELENIUM	4	-	-	< 5
SULFATE *	4	100 ± 36	81	137
TEMPERATURE **	4	11.7	10.0	13.2

 *
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

TABLE 41
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 7A
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	4	-	-	< 5
BARIUM	4	157 ± 88	72	225
CHLORIDE *	4	31 ± 20	18	49
DISSOLVED SOLIDS *	4	686 ± 145	589	829
FLUORIDE	3	126 ± 29	108	138
IRON	4	224 ± 346	100	598
MANGANESE	4	340 ± 60	299	400
MERCURY	4	-	-	< 0.05
PH	4	-	7.0	7.2
SELENIUM	4	-	-	< 5
SULFATE *	4	146 ± 31	114	165
TEMPERATURE **	4	11.5	10.0	14.1

*
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

TABLE 42
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 5
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	74 ± 53	61	86
CHLORIDE *	2	9 ± 4	8	10
DISSOLVED SOLIDS *	2	350 ± 228	297	403
FLUORIDE	1	-	-	158
IRON	2	-	-	< 100
MANGANESE	2	156 ± 557	26	285
MERCURY	2	-	-	< 0.05
PH	2	-	7.6	7.9
SELENIUM	2	-	-	< 5
SULFATE *	2	119 ± 120	91	147
TEMPERATURE **	2	10.3	9.7	10.9

*
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

TABLE 43
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 HELL 8
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	107 ± 92	86	128
CADMIUM	2	0.50 ± 0.86	0.30	0.70
CHLORIDE *	2	59 ± 28	52	65
COPPER	2	8 ± 18	4	12
DISSOLVED SOLIDS *	2	736 ± 39	727	745
FLUORIDE	1	-	-	210
IRON	2	-	-	< 100
LEAD	2	1 ± 1	1	1
MANGANESE	2	92 ± 3	91	93
MERCURY	2	-	-	< 0.05
NICKEL	2	26 ± 16	22	29
PH	2	-	7.3	7.3
SELENIUM	2	-	-	< 5
SILVER	2	-	-	< 0.20
SULFATE *	2	129 ± 32	121	136
TEMPERATURE **	2	11.5	10.8	12.3
ZINC	2	15 ± 22	10	20

*
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

TABLE 44
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 9
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	9 ± 19	< 5	14.0
BARIUM	2	227 ± 476	116	338
CADMIUM	2	0.65 ± 0.22	0.60	0.70
CHLORIDE*	2	165 ± 43	155	175
COPPER	2	11 ± 2	10	11
DISSOLVED SOLIDS*	2	1090 ± 110	1070	1120
FLUORIDE	1	-	-	212
IRON	2	1390 ± 2870	730	2060
LEAD	2	1 ± 1	1	1
MANGANESE	2	3250 ± 2570	2650	3840
MERCURY	2	0.10 ± 0.22	< 0.05	0.15
NICKEL	2	41 ± 19	37	45
PH	2	-	6.6	6.7
SELENIUM	2	-	-	< 5
SILVER	2	< 0.20	< 0.20	0.30
SULFATE*	2	41 ± 54	28	53
TEMPERATURE**	2	12.3	11.2	13.5
ZINC	2	15 ± 22	10	20

*
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

TABLE 45
 CHEMICAL CONSTITUENTS AT ANL LANDFILL
 WELL 10
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	83 ± 2	83	83
CADMIUM	2	-	-	< 0.20
CHLORIDE *	2	20 ± 9	18	22
COPPER	2	4 ± 13	1	7
DISSOLVED SOLIDS *	2	575 ± 349	494	656
FLUORIDE	1	-	-	248
IRON	2	-	-	< 100
LEAD	2	1 ± 1	1	1
MANGANESE	2	567 ± 118	540	595
MERCURY	2	-	-	< 0.05
NICKEL	2	22 ± 6	20	23
PH	2	-	7.4	7.4
SELENIUM	2	-	-	< 5
SILVER	2	-	-	< 0.20
SULFATE *	2	143 ± 19	138	147
TEMPERATURE **	2	11.8	10.1	13.5
ZINC	2	15 ± 22	10	20

*
 CONCENTRATION IN MILLIGRAM/LITER.

**
 DEGREES CENTIGRADE

Samples for organic analyses were collected in serum bottles which were immediately sealed with Teflon-lined septa using a crimping tool. The samples were refrigerated until analysis. The samples were extracted with *n*-pentane and analyzed by gas chromatography with mass spectrophotometric detection. The mass spectra were acquired using pre-selected masses which increases sensitivity and provides for scanning of compound groups such as halogenated hydrocarbons and benzene derivatives. Results are shown in Table 46. Compounds detectable by this system were not observed. All values are corrected for recoveries obtained from duplicate samples with known amounts of each compound added. One duplicate was analyzed with each set. The tests were performed to determine whether selected materials were moving from the burial site and any positive results are considered significant. As a point of reference, however, organic levels of greater than 50 µg/L can be considered significant health hazards from materials such as benzene. For other materials included in this study such as toluene, the levels which pose health risks are significantly higher.

TABLE 46
ORGANIC CONSTITUENTS AT ANL LANDFILL

ORGANIC MATERIALS (MICROGRAMS/LITER)	WELL LOCATIONS												
	1	1-2	2	2-2	3	4	4-2	5	6	7A	8	9	10
BENZENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
ETHYLBENZENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
M-DICHLOROBENZENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
M-XYLENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
MONOCHLOROBENZENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
O-DICHLOROBENZENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
O-XYLENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
P-DICHLOROBENZENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
P-XYLENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
PERCHLOROETHYLENE	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
TOLUENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
TRICHLOROETHYLENE	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
1,2,4-TRICHLOROBENZENE	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5

FOUR SAMPLES WERE COLLECTED AT LOCATIONS 3, 6, AND 7A. TWO SAMPLES WERE COLLECTED AT THE OTHER LOCATIONS.

IV. APPENDIX

A. References

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B. Environmental Permits, Impact Statements, and Activities

The environmental permits in effect at ANL are shown in Table 47. As shown, all the permits have been issued by the Illinois Environmental Protection Agency (IEPA). The compliance status of the NPDES permit is discussed in Section III.B.1. The monitoring data obtained for the landfill are discussed in Section III.B.2. The limits or standards are not specified in the permit to operate the landfill, but the number of monitoring wells and the types of measurements and results are within the IEPA guidelines.

For Boiler No. 5 operations, sulfur dioxide and particulates are measured in the exhaust stack and the results reported to the IEPA. The Boiler is operated in compliance with the permit.

TABLE 47

IEPA Environmental Permits in Effect at ANL

Permit Type	Facility	Permit/ Application No.	Expires
Fossil Energy Users Laboratory (FEUL) Facility Air Emissions	FEUL-MHD Building 145	ID 043802AAA App. C8012024	9/13/87
Operating	Steam Plant - Boiler No. 5 Building 108	ID 043802AAA D/02 App. 79090047	8/1/88
Construct and Own	Water pollution control facility, coal-pile run- off containment area Building 108	1980-EB-1568	Covered by NPDES Permit 3/1/89
Construction Operation	Emco Wheaton coaxial recovery system for gas dispensing faci- lities - Building 827	HG490	5/30/90
NPDES	Laboratory Water Effluents	IL0034592	3/1/89
Sludge Drying Bed Replacement	Sanitary Plant Laboratory Plant Building 570	1981-EB-1776	Covered by NPDES Permit 3/1/89
Lift Station Construction	Boiler waste-water, coal-pile runoff over- flow, lime pond and water pond overflow Building 108	1982-HB-0402	Operation covered by NPDES Permit 3/1/89
Operating	Landfill 800 Area	1981-20-0P	Special Conditions
RCRA Hazardous Waste Storage, Generation, and Treatment	Facilities 306, 317, 325A, 325C, and 329	IL3-890-008946	Interim
Methanol Storage Tank	Coaxial Vapor Control System Building 827	App. 86020043	2/7/91

The vapor recovery system for gasoline and alcohol vehicle fuel dispensing was constructed, and is operating, as designed in accordance with the permit.

The environmental effects of new construction projects, modifications to facilities, and other activities that can have an environmental impact are evaluated before work is begun. During 1986, no draft or final Environmental Impact Statements or Environmental Assessments were completed, and no significant pollution abatement projects were undertaken.

C. Environmental Quality Standards, Detection Limits, Analytical Errors, and Quality Assurance

1. Standards

All of the standards and detection limits for chemical constituents, and some of those for radionuclides and external radiation, are given in the main body of the report. The standards used in this report for determining the population exposure are the EFF.D.E. factors provided in the DOE interim guidance.⁶ The applicable EFF.D.E.s for calculating the committed dose due to inhalation and ingestion exposure are given in Table 24, Section III.A.7. Except for gaseous emissions, where the chemical form was generally known, the most restrictive EFF.D.E. was selected when more than one choice was available.

2. Detection Limits

The detection limits were chosen so that the measurement uncertainty at the 95% confidence level is equal to the measured value. In Table 48 are collected air and water detection limits (minimum detectable amounts) for all radionuclides, and for those materials, for which measurements were made. The relative error in a result decreases with increasing concentration. At a concentration equal to twice the detection limit, the error is about 50% of the measured value, and at 10 times the detection limit, the error is about 10%.

TABLE 48

Detection Limits
($\mu\text{Ci/mL}$)

Nuclide or Activity	Detection Limit	
	Water	Air
Americium-241	1×10^{-12}	-
Beryllium-7	-	5×10^{-15}
Californium-249	1×10^{-12}	-
Californium-252	1×10^{-12}	-
Cesium-137	-	1×10^{-16}
Curium-242	1×10^{-12}	-
Curium-244	1×10^{-12}	-
Hydrogen-3	1×10^{-7}	-
Neptunium-237	1×10^{-12}	-
Plutonium-238	1×10^{-12}	3×10^{-19}
Plutonium-239	1×10^{-12}	3×10^{-19}
Radium-226	1×10^{-10}	-
Strontium-89	2×10^{-9}	1×10^{-16}
Strontium-90	2.5×10^{-10}	1×10^{-17}
Thorium-228	-	1×10^{-18}
Thorium-230	-	1×10^{-18}
Thorium-232	-	1×10^{-18}
Uranium-234	-	1×10^{-19}
Uranium-235	-	1×10^{-19}
Uranium-238	-	1×10^{-19}
Uranium - natural	2×10^{-10}	2×10^{-17}
Alpha	2×10^{-10}	2×10^{-16}
Beta	1×10^{-9}	5×10^{-16}

3. Quality Assurance Program

a. Radiochemical Analysis and Radioactivity Measurements

All nuclear instrumentation is calibrated with standard sources obtained from the NBS, if possible. If NBS standards were not available for particular nuclides, NBS traceable standards from the Amersham Corporation were used. The equipment is usually checked on a daily basis with secondary counting standards to insure proper operation. Samples were periodically analyzed in duplicate or with the addition of known amounts of a radionuclide to check precision and accuracy. In addition, standard and intercomparison samples distributed by the Quality Assurance Branch of the U. S. Environmental Protection Agency (EPA-QA) are analyzed regularly. Results of our participation in the EPA-QA program during 1986 are given in Table 49. In the table, the comparison is made between the EPA-QA value, which is the quantity added to the sample by that Laboratory, and the value obtained in our laboratory. To assist in judging the quality of the results, typical errors for our analyses are 2% to 50% depending on the concentration and the nuclide, and the errors in the EPA-QA results are 2% to 5% (our estimate).

In addition, participation continued in the DOE Environmental Measurements Laboratory Quality Assurance Program (DOE-EML-QAP), which for 1986 was a single distribution of four or five different sample matrices containing various combinations of radionuclides. Results for 1986 are summarized in Table 50. In the table, the comparison is made between the EML value, which is the result of replicate determinations by that Laboratory, and the value obtained in our laboratory. To assist in judging the quality of the results, typical errors for our analyses are 2% to 50% and the errors in the EML results are 1% to 30% (depending on the nuclide and the amount present). For most analyses for which the differences are large (> 20%), the concentrations were quite low and the differences were within the measurement uncertainties.

TABLE 49

Summary of EPA-QA Samples, 1986

Type of Sample	Analysis	Number Analyzed	Average Difference from Added
Air Filter	Total Alpha	2	35%
	Total Beta	2	3%
	Strontium-90	2	7%
	Cesium-137	2	38%
Milk	Potassium-40	2	4%
	Strontium-89	1	3%
	Strontium-90	1	3%
	Iodine-131	3	21%
	Cesium-137	2	20%
Water	Total Alpha	1	56%
	Total Beta	1	22%
	Hydrogen-3	3	3%
	Chromium-51	2	4%
	Cobalt-60	4	2%
	Zinc-65	3	3%
	Strontium-89	3	6%
	Strontium-90	3	6%
	Ruthenium-106	2	16%
	Iodine-131	2	12%
	Cesium-134	4	8%
	Cesium-137	4	13%
	Radium-226	2	31%
	Radium-228	2	12%
	Total Uranium	3	7%
Plutonium-239	2	10%	

TABLE 50

Summary of DOE-EML-QAP Samples, 1986

	Difference From EML Value				
	Air Filters	Water	Soil	Tissue	Vegetation
Hydrogen-3	-	1%	-	-	-
Beryllium-7	6%	-	-	-	-
Potassium-40	-	-	2%	34%	10%
Manganese-54	2%	6%	-	-	-
Cobalt-60	5%	3%	-	-	-
Strontium-90	0%	2%	10%	-	8%
Cesium-137	2%	0%	1%	-	5%
Radium-226	-	-	8%	40%	-
Uranium-234	1%	9%	36%	-	-
Uranium-238	1%	9%	31%	-	-
Plutonium-238	-	-	2%	-	4%
Plutonium-239	0%	3%	4%	-	4%
Americium-241	2%	17%	-	-	44%

b. Penetrating Radiation

Our laboratory participated in the Eighth International Environmental Dosimeter Intercomparison Project conducted by the DOE Radiological and Environmental Sciences Laboratory in Idaho Falls, Idaho, and the DOE Environmental Measurements Laboratory (EML) in New York, New York. Participants supplied four sets of each type of dosimeter they wished to evaluate. Two sets were exposed in the field to a mixture of natural and man-made gamma radiation; one set was exposed in the laboratory to a known (but unrevealed) cesium-137 source and the fourth set served as a control. After exposure, the dosimeters were returned to the participant for measurement. The field exposure was measured by an EML high-pressure ion chamber for comparison. The results, in milliroentgens (mR), were:

	<u>Field Site #1</u>	<u>Exposure</u> <u>Field Site #2</u>	<u>Laboratory</u> <u>(Cs-137)</u>
Delivered Dose	29.7 \pm 1.5	10.4 \pm 0.5	17.2 \pm 0.9
ANL Value	28.9 \pm 6.2	10.1 \pm 4.5	16.2 \pm 3.4

The uncertainties listed are the standard deviations as estimated by the sponsors for their values and by us for our results. All results agree with the delivered dose within the measurement error at the 95% confidence levels. The most important comparison is for the field exposures. Here the differences between the ion chamber result and our results were 0.8 mR \pm 7.7 mR and 0.3 mR \pm 5.0 mR, respectively. The standard deviations for our results are relatively large because the transit and storage dose, as measured with the control dosimeters, is significant and must be subtracted from the total dose.

c. Chemical Analyses

Since July 1, 1985, all work has been performed in accordance with the Quality Assurance Plan (QAP) for the Industrial Hygiene Chemistry Laboratory (H0030-0002-QA-00). This QAP specifies quality-related activities that are necessary to obtain the required results with the required accuracy and precision.

All instrumentation is calibrated with NBS standard sources when available. Samples from the EPA are also used for these purposes. Duplicate analyses are performed frequently to check precision. Known amounts of standard materials are also routinely analyzed to measure accuracy.

The laboratory participates in the National Institute of Occupational Safety and Health (NIOSH) Proficiency Testing Program (PAT) which requires analyses of many materials of environmental interest. Results are in good agreement with the NIOSH values and are listed in Table 51.

TABLE 51

Summary of NIOSH-PAT Program Samples, 1986

Constituent	Average Difference From Reference Value
Cadmium	2.2% (16)
Lead	2.1% (16)
Zinc	7.6% (16)
Benzene	5.3% (4)
Carbon Tetrachloride	3.6% (8)
Cellosolve Acetate	7.0% (4)
Chloroform	3.2% (4)
Dichloroethane	6.3% (4)
p-Dioxane	4.6% (4)
Toluene	4.8% (4)
Trichloroethylene	5.3% (4)
O-xylene	5.9% (8)

Note: The figures in parentheses are the number of samples analyzed.

The laboratory also participates in the U. S. Environmental Protection Agency Discharge Monitoring Report Quality Assurance Program (EPA-DMR-QAP). Results were rated acceptable by the EPA and appear in Table 52.

TABLE 52

Summary of EPA-DMR-QAP Non-Radiological Samples, 1986

<i>Constituent</i>	<i>Average Difference From Reference Value</i>
<i>Mercury</i>	<i>5%</i>
<i>Zinc</i>	<i>6%</i>
<i>pH</i>	<i>0</i>
<i>Total Suspended Solids</i>	<i>15%</i>
<i>Oil and Grease</i>	<i>3%</i>
<i>Chemical Oxygen Demand</i>	<i>3%</i>
<i>Biochemical Oxygen Demand</i>	<i>13%</i>

d. Sampling, Sample Storage, Other

Many factors enter into an overall quality assurance program other than the analytical quality control discussed above. Representative sampling is of prime importance. The continuous water sampler in Sawmill Creek provides a representative sample for a critical sampling location since the concentration of pollutants in the waste water may vary appreciably during each 24-hour period.

The accuracy of the flowmeters in the air sampling equipment is verified periodically with a calibrated rotameter, as well as by comparison to a Roots meter primary standard.

Samples are pre-treated in a manner designed to maintain the integrity of the constituent sought. For example, samples for trace radionuclide analysis are acidified immediately after collection to prevent hydrolytic loss of metal ions.

D. Acknowledgements

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